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Review of the Status of Cumulative Fission Yields from 239Pu(n,f) of Interest to Nuclear Forensics

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Review of the Status of Cumulative Fission Yields from ^{239}Pu(n,f) of Interest to Nuclear Forensics

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Report Summary

The cumulative fission yields of the four nuclides ^{95}Zr, ^{99}Mo, ^{144}Ce and ^{147}Nd are key parameters in estimating the effects from nuclear tests and for post-detonation nuclear forensics in general. Driven particularly by differences in the fission yields from ^{239}Pu(n,f) that have been in use by LLNL and LANL over the years, an intensive effort has been undertaken to evaluate the current state of our knowledge of these yields and to provide a basis for projecting yields to those applicable to forensics. The neutron spectrum producing fission in the Jezebel critical assembly ($< E_n > \sim 1.9$ MeV) may be taken as a reasonable reference. It is desired to know the cumulative fission yields to a total uncertainty (1σ) of ± 2.5%.

Of critical importance to approaching this uncertainty goal is the determination of the possible energy dependence of fission yields in the mass ranges $90 \leq A \leq 100$ and $143 \leq A \leq 154$. To date all available evidence suggests that the energy dependencies of the yields of ^{95}Zr, ^{99}Mo, ^{144}Ce and ^{147}Nd must be smaller than or comparable to the uncertainty of ~ 4-5% that has been typical of an individual fission yield determinations with high-quality radiochemical methodology. As a result, we have examined in some detail data acquired through isotope dilution methodology applied to Pu targets irradiated in various fast reactor environments. Although limited to average neutron energies $< E_n > \leq 1.3$ MeV, the isotope dilution methodology has provided isotopic abundances for a number of fission product elements with reported uncertainties as small as ± 0.25% and absolute fission yields with typical uncertainties of ~ ± 2%. These measurements afford an approach to examination of the energy dependence of fission yields with the lowest uncertainties of any approach thus far reported in the literature.

The relative isotopic abundances of Nd isotopes $^{143\text{-}146}$Nd and ^{148}Nd, which can be considered chain yield monitors, display systematic variations with average neutron energy and mass number. The scatter in the data is relatively small and the existence of energy dependence in some chain yields is very strong. Given the near constancy of the elemental yield in Nd, the data imply energy dependence of the yields for $A \geq 146$ and for $A \leq 144$. When cumulative yield data for the Nd isotopes are combined with the cumulative yield data for the Sm isotopes with A = 147, 149, 151, 152 and 154, systematic variations with energy and mass number are clearly evident and are completely consistent with the variations seen in the relative abundance data despite an increased estimated uncertainty of ~ 2%. Taken together these data indicate energy dependence of the fission yield of ^{147}Nd over the energy range $< E_n > \leq 1.3$ MeV. However, the data do not permit an estimate of the magnitude of the ^{147}Nd yield at $< E_n > = 1.9$ MeV with any reasonable certainty.

The elemental yield of Zr from the experiments performed at the Idaho National laboratory, the only such measurements by isotope dilution methodology reported to date, displays an approximate linear increase with energy but there is considerable scatter in isotopic abundances measured for A = 91 - 94 and 96. On the other hand, both the elemental and isotopic abundances of the Mo isotopes of A = 95, 97, 98 and 100 are essentially constant over the energy range $< E_n > \leq 1.3$ MeV. It is not possible to judge

the consistency of the Zr and Mo data with these limited data. One can conclude that any energy dependence of ^{95}Zr and ^{99}Mo over the energy range probed is likely to be small (≤ 2%).

Comparison of the projections of the energy variations of the cumulative yields of ^{95}Zr, ^{99}Mo, ^{144}Ce and ^{147}Nd from our analysis of the isotope dilution experiments with the most current evaluations of LLNL and LANL, assuming the latter correspond to an average neutron energy of $< E_n > = 1.5$ MeV, indicates consistency within experimental errors (~ 3%).

We conclude that, although all of the most advanced evaluations are in agreement, it is not possible to project cumulative yields to the neutron spectrum producing fission in the Jezebel critical assembly ($< E_n > \sim 1.9$ MeV) with confidence. Projections are fraught with error because the functional form of the energy dependence is unknown. Despite this caveat, several simple projections are made and in the case of ^{147}Nd, the range in median values is 2.11% - 2.16%. For the remaining isotopes, it is not possible to exclude observation of an energy dependence that is consistent with the goal of uncertainties of ± 2.5% without additional and highly accurate experimentation.

The goal of achieving experimental cumulative fission yields as small as desired at elevated neutron energies is essentially unprecedented. As a result, we feel that a theoretical effort in combination with experiments performed with multiple techniques offers the best path forward to establish the parameters for the energy dependence of the specific nuclide yields of direct interest. A theoretical parameterization of the energy dependence is required to make use of limited data and, at the same time, to provide guidance to additional energy regimes as may prove of interest.

There is little doubt that much better experimental data must be acquired with a design goal of attaining absolute uncertainties of about 1% if the goal of an absolute uncertainty of ± 2.5% is to believed in the ever present concern over unknown systematic errors. In particular, cumulative fission yields obtained by essentially all methodologies are limited by the uncertainties in the determination of the number of fissions that have taken place in a target under study. This problem must be a focus of study in the future.

We propose that the national laboratories develop an intense, variable energy mono-energetic neutron source for measurement of the chain yields as a function of energy. We further propose that a combination of established radiochemical techniques and mass spectrometry, along with exploration of new experimental studies to reduce the principal uncertainties that are known to exist, be implemented in the near future. Promising methodologies such as time projection chamber technology and direct reaction coincidence techniques should be explored. Such a broad approach has the potential of providing accurate results with relatively independent methodologies and thereby lessens the likelihood of systematic limitations. Finally, the establishment of energy-dependent standards with proven uncertainties of ± 1% or less can be used to normalize past results from radiochemical measurements and can be used to normalize data obtained in future reactor-based or critical assembly-based measurements.

We are struck by the fact that the problem of accurate fission chain yields at elevated neutron energies has languished for something like three decades, despite the frequent meetings and discussions between the personnel at LLNL and LANL. We recognize that the efforts at the laboratories are guided by programmatic dictates to a large extent; nevertheless, this problem is clearly one that is fundamental to nuclear forensics and should have raised interest in the scientific community.

We cannot help but feel that a requirement for publication of purely scientific information in peer-reviewed journals of the highest quality would have led to the type of intense study now taking place and might well have solved the problem at a much earlier date. We cannot stress the importance of peer-reviewed publications strongly enough. It is in the best interests of the institutions and the nation.

I. Introduction

On June 12 2009, we were asked to form a small committee to provide expert opinion on the status of the cumulative fission yields of selected nuclides of importance to the nuclear weapons program and nuclear forensics. This committee was to be completely independent of two large groups within LLNL and LANL charged with review of all available data in an attempt to understand historical differences in the yields used by the two laboratories that affect the interpretation of test data.

The committee began its work toward the end of June 2009 and the finalized charge to the committee is as follows.

> As indicated in the document **Proposed Panel Review: Joint LANL/LLNL Fission Product Evaluation** (PPR) both laboratories are currently engaged in an intensive study to resolve historic differences in fundamental nuclear data sets. Among other things, this data is used to assess device performance through radiochemical evaluation of device debris. Current estimates of the uncertainties desired in fission yields are approximately $1\sigma = \pm 2.5\%$.
>
> As part of this process, LLNL has asked a completely Independent Panel (IP) of experts to provide a scientifically rigorous analysis of the quality of fission yields and an evaluation of the best fission chain yield for the select set of fission products specified in the PPR. A report from the IP is required by LLNL by the end of October 2009.
>
> Because of the short time available, the scope of the IP's work should be limited primarily to assessing the current state of fission yields for ^{239}Pu under irradiations by both thermal and fission spectrum neutrons. With respect to the latter, it is the irradiation of ^{239}Pu in the metallic state that is of direct concern where the sources are prompt-fission neutrons.
>
> The effort of the IP should be directed to those publications and reports deemed to represent state-of-the-art experiments in both design and execution and where sufficient information is available to rigorously judge both systematic and statistical errors. Because of the range of methodologies that are employed in fission-yield measurements, concerns may be raised that cannot be addressed by the IP with high confidence. In such a case, the IP is requested to engage the expertise of any national or international expert who might provide the necessary information.
>
> The IP's report should include judgments concerning the quality of the best available experimental data and the science behind the experiments, as well as concerns with respect to any effects that tend to raise significant questions concerning the uncertainty estimates provided. If the quality of all or specific fission yields are suspect, suggestions of additional experiments that might provide significantly reduced uncertainties should be discussed. The evaluated

fission chain yield results should represent the best evaluation possible under the most rigorous circumstances.

Any comments the IP might also have on the validity of "evaluations" or "translational methods" of fission-yield data to generate "new or corrected data" would also be helpful.

The nuclides of central interest were enumerated in the charge to the LLNL and LANL review committees by C. P. Verdon and C. F. McMillan. They are ^{147}Nd (primary) and ^{95}Zr, ^{99}Mo, ^{144}Ce, ^{155}Eu and ^{156}Eu secondary (LLNL) and ^{147}Nd and ^{99}Mo (primary) and ^{95}Zr, ^{140}Ba and ^{144}Ce (secondary) (LANL).

II. Philosophy and General Guidelines Used by the Committee

The desire to know cumulative fission yields to a 1σ uncertainty of ± 2.5% in the irradiation of ^{239}Pu with a neutron spectrum characteristic of a metallic critical assembly is perhaps unprecedented. Other than the intensive studies of ^{235}U and, secondarily, ^{239}Pu thermal fission over many years, there are but few studies in the open literature that have reported measurements with such uncertainties at elevated neutron energies. As a result the committee sought the most accurate information available with the following guidelines:

1. Only primary data sources were considered if at all possible. These include publications in learned journals and technical reports. Evaluated data were not considered except for general guidance and assessment of average trends in experimental data.

2. Effort was concentrated on those publications and reports that contained the majority of the information needed to judge the overall quality of the data presented, including

a. The general methodology, details of the individual procedures, chemical procedures and tests performed to ensure their adequacy.
b. The means by which confounding issues (interferences, calibrations, etc.) were treated.
c. The means by which the absolute number of fissions were determined.
d. The means by which random and systematic errors were considered and combined to provide an overall uncertainty estimate.

While the quality of data obtained by the radiochemistry groups at LLNL and LANL appears to be excellent, the majority of written reports available to us did not contain sufficient information to fulfill many of the requirements listed above. As a result we have not considered these data in our primary effort. This should provide as independent an appraisal of the current state of knowledge of cumulative fission yields as our committee can provide.

Accurate determination of the cumulative yield of fission products is clearly a difficult experimental issue. We have attempted to consider not only the status of the best experimental data but also the theoretical issues that might affect and confound their interpretation. In the following both are addressed along with a discussion of possible routes by which the desired accuracy might be achieved in new state-of-the art experiments and theoretical studies.

In addressing the current status of fission yields, we have taken the neutron energy spectrum of the critical assembly Jezebel as the reference standard for which high-quality data are required. The spectrum producing fission in this assembly has an average energy of about 1.9 MeV. As a result, it is important to recognize that the majority of fission yields have been performed with neutron spectra of considerably reduced average energy. All experiments described in the open literature that report the most accurate cumulative fission yields were performed in fast reactors with average neutron energies less than ~ 1.3 MeV. Most irradiations performed with critical assemblies involved average neutron energies below about 1.5 MeV.

III. Experiments Considered

1. General

The most accurate measurements of cumulative fission yields have resulted from complex and highly-sophisticated methodologies developed over decades of research and development. They require extensive expertise and great care in their performance. We have not studied all of these but believe it appropriate to provide some general comments concerning the principal methodologies of "classical" radiochemistry and isotope dilution mass spectrometry and the principal sources of uncertainties attendant to each.

Classical radiochemistry depends primarily upon determination of the number of atoms or disintegration rate of a fission product by measurement of its radioactive decay through assay of one or more γ rays or the β particles emitted in its decay along with measurement of the number of fissions produced in an experiment. The main uncertainties arise from determination of the number of fissions, the yields of the chemical separations required for isolation of a fission product element, the efficiencies of the radiation detectors employed and, when γ rays are assayed, uncertainties in absolute photon abundances.

The most sophisticated modern approach to determining the number of fissions is through use of a fission chamber containing a small mass of the fissile nuclide and relating this to the number of fissions induced in a larger mass from which a fission product is isolated, both masses being irradiated simultaneously. The relative masses of the targets must be known accurately and the magnitude and shape of the neutron spectrum producing fissions in the two targets must also be accurately known.

The most sophisticated approaches to assay of a fission product rely on minimization of uncertainties by performing a measurement relative to one or more reference nuclides for which the cumulative fission yield has been determined with high accuracy. The nuclide ^{99}Mo has been the reference historically used at LANL.

The isotope dilution approach relies on the ability to accurately determine the relative abundances of multiple isotopes of an element in a well-calibrated mass spectrometer with two aliquots of an irradiated target. One aliquot contains only the atoms of a fission product element and the second contains, in addition, an accurately known number of atoms of one of the isotopes of the product element. So long as both aliquots represent accurate fractions of the total target, the absolute number of atoms of a fission product can be determined with high accuracy by solving a simple set of linear equations.

In theory, the absolute number of fissions in an isotope dilution experiment can be determined by measurement of the total number of fission product atoms for all mass chains in one peak from binary fission. In practice this limit cannot be achieved but some 80 - 90% or more of the total atoms in the heavy-massed peak have been measured in many cases. To account for atoms in unmeasured mass chains, two approaches have been used. In the first, the most accurate evaluated fission yields for the missing mass chains are taken to represent the fraction of fissions not measured. In the second, interpolation schemes are used between measured mass chains with the assumption of a smooth continuous mass yield distribution function.

A second means for determining the total number of fissions produced in a target is to measure the difference in mass of the fissile nuclide before and after irradiation. In practice, this method appears to have produced inferior results compared to that discussed above.

The principal sources of uncertainty in fission yield determinations via isotope dilution experiments arise from the introduction of atoms of the element of interest during target dissolution and chemical separation procedures, the presence of contamination from species with the same mass numbers as the fission product isotopes assayed, corrections for the fraction of the total fission product atoms not directly measured and, in some cases, corrections for neutron reactions that affect the observed atoms of a specific nuclide.

It should be emphasized that both the classical radiochemical and isotope dilution approaches rely on high-quality radiochemical separations of the elements of interest although the need for extensive decontamination from interfering nuclides varies considerably with the methodology employed and the specific fission product of interest. An exception to this is the use of high-resolution γ-ray spectroscopy, which can, in many cases, provide quantitation of all the major fission products in the single sample without the need for chemical separations.

2. Experiments Considered from the Open Literature

With but one exception, the experiments chosen for detailed study are all based on isotope dilution methodology. First and foremost, this methodology provides as its primary result the relative isotopic abundances of fission product elements with very small uncertainties. For those elements with a significant number of stable or long-lived isotopes, it is then possible to use these data directly for a relatively sensitive search for the possible energy dependence of fission yields over a restricted range in mass number. Second, the methodology permits direct measurement of the majority of the total fission yield in the heavy-massed peak. When combined with reasonable estimates of the yield of mass chains not directly measured, these measurements provide absolute fission yields with overall uncertainties that tend to be about one-half those reported from radio-chemical methodologies.

The sources of cumulative fission yields measured with isotope dilution methodology considered here are as follows:

a. W.J. Maeck et al., "Absolute Thermal Fission Yields for ^{239}Pu" [1] - report on thermal fission yields measured at the Idaho National Laboratory from irradiations in the Engineering Test Reactor. The abundance of ^{239}Pu in the targets was about 0.991. The irradiation was conducted in the reflector region of the core ensuring a reasonably well-moderated thermal neutron distribution. The capture-to-fission ratio was about 0.46 - 0.48. Absolute cumulative fission yields were determined by use of evaluated data to account for unmeasured mass chains.

b. W.J. Maeck, ed., "Fast Reactor Fission Yields for ^{239}Pu and ^{241}Pu" [2] - report on fission yields from irradiation in EBR-II Row 8. The abundance of ^{239}Pu was about 0.991. The average neutron energy indicated by Maeck was in the range 0.2 - 0.5 MeV depending on the location of an individual specimen capsule in the reactor. Absolute cumulative yields were determined by means of an interpolation scheme to account for unmeasured mass chains.

c. W.J. Maeck, A.L. Erikson and R.L. Tromp, "Fast Reactor Fission Yields for ^{241}Pu and Relative Isotopic Data for ^{239}Pu Irradiated in Row-4 of EBR-II" [3] - the abundance of ^{239}Pu in the target was 0.994. The irradiation was actually carried out in Rows 2 and 4, each for about the same total neutron fluence. Consideration of the core geometries suggested that the energy distributions at the two irradiation sites should have been similar but this was not proven experimentally. The average neutron energy in the irradiation was estimated to be ~ 700 keV by the authors. An estimated average energy of ~ 730 keV is found from the measured cumulative yield ratio ^{150}Nd/^{143}Nd and the correlation shown in Figure 1 of ref. 4. Only relative isotopic abundances of various fission product elements were reported because of incomplete dissolution of the fuel specimen. The authors stated that there was no indication of any effects that could have significantly influenced the isotopic abundances of the elements in the dissolved fraction of the sample.

d. F.L. Lisman et al., "Fission Yields of Over 40 Stable and Long-Lived Fission Products for thermal Neutron Fissioned ^{233}U, ^{235}U, ^{239}Pu and Fast Reactor Fissioned ^{235}U and ^{239}Pu"[5] - thermal neutron irradiations were performed in the Materials Testing Reactor at the National Reactor Testing Station. Fast fission irradiation was carried out with a special ^{239}Pu-fueled rod irradiated in EBR-I. The abundance of ^{239}Pu was 0.992. The average neutron energy in the irradiation was estimated to be ~1330 keV from the measured yield ratio ^{150}Nd / ^{143}Nd and the correlation of Figure 1 of ref. 4. Absolute chain yields were obtained primarily by interpolation between measured mass chains and extrapolations assuming a smooth and continuous mass yield distribution function.

e. W. Davies, "Absolute Measurements of Fission Yields for 235-Uranium and 239-Plutonium in the Dounreay Fast Reactor" [6] - the abundance of ^{239}Pu in the fuel was 0.939. The authors report that the neutron energies were in the range 10.9 keV $\leq E_n \leq$ 2.23 MeV. The mean neutron energy estimated from the reported yield ratio $^{150}Nd/^{143}Nd$ and the correlation given in Figure 1 of ref. 4 is ~ 720 keV. Absolute cumulative yields were obtained by measurement of the burnup of the fuel with an estimated uncertainty in this quantity of ± 4.8%.

f. V.Ya. Gabeskiriya et al., "Fission Yields from ^{235}U and ^{239}Pu Irradiated in the BOR-60 Reactor" [7] - this is a very brief report without extensive detail on the experimental methods and procedures. The abundance of ^{239}Pu in the fuel is not given but the authors provide a spectral index in terms of the ratio of the fission cross sections ^{238}U / ^{235}U. The average neutron energy estimated from the yield ratio ^{150}Nd / ^{143}Nd and the correlation given in Figure 1 of ref. 4 is ~ 735 keV. Absolute cumulative yields were obtained by setting the sum of the relative yields of ^{145}Nd and ^{146}Nd to the sum of the absolute yields reported by Lisman et al.[5] and thus the absolute fission yields reported here are correlated to the Lisman et al. results.

g. L. Koch, "Status Report on Fast Reactor Fission Yields in the TACO Experiment" [8] - this is a brief interim report on irradiations performed in the Rapsodie fast reactor. The composition of the Pu target was not given. Only total cumulative fission yields were reported and thus the directly-measured relative isotopic abundances were not available to us. An average neutron energy of ~ 594 keV was derived from the reported ^{150}Nd / ^{143}Nd yield ratio using the correlation given in Figure 1 of ref. 4.

We note that the reports of the last three references contain far less information on the details of the experiments, data analysis, etc., than those of the preceding four reports. We include the data from the last three primarily because of the smallness of the data set and because they serve as a means for partially assessing the presence or absence of significant systematic errors from the experiments described in the first four references.

In addition to these reports, we have studied the work by Laurec and co-authors, the most important of which is that of ref. 9. It provides extensive details on the experimental and data analysis effort on fission yield determinations from irradiations in critical assemblies and from 14-MeV neutron irradiations by a radiochemical approach. Due to our interest

in studying the energy dependence of fragment yields the focus of our effort has been on the isotope dilution measurements and so we restrict ourselves to some general comments about the Laurec results and the need for more documentation of LLNL measurements.

Laurec et al. published radiochemical cumulative yield measurements from irradiation of ^{233}U, ^{235}U, ^{238}U, and ^{239}Pu with fission spectrum and 14.7 MeV neutrons. The abundance of ^{239}Pu in the target used for the fission spectrum measurements was 0.983. Absolute determination of the number of fissions was obtained from a fission chamber. Although Monte Carlo simulations of the neutron spectra at various locations in the two critical assemblies used are presented, no experimental measurements on the neutron spectra are reported. According to the authors, the ^{239}Pu irradiations were performed along with the ^{235}U irradiations.

While the majority of the measurements reported by Laurec et al. are in excellent agreement with other measurements, there is a fairly large discrepancy between the two for the absolute cumulative yields reported from irradiation of ^{239}Pu with fission spectrum neutrons. Although the most precise results were reported with uncertainties of about 3.7%, the ratios of fission product activities, that exclude uncertainties in nuclear data constants and detector efficiencies, had uncertainties in the range 0.3 - 0.6%. Accurate and precise absolute chain yields require that the total number of fissions, target masses and detector efficiencies be known accurately and precisely, and flux corrections must be determined accurately. Laurec reported uncertainties in the determination of the total fissions in a sample of ~ 0.5%, in normalization of target masses of ~1-2%, in normalization of detector efficiencies of ~1.5%, and in flux normalization of ~ (0 -1.5)%. If these values are correct, the discrepancy between the Laurec cumulative yields and those from other measurements (especially Maeck) is puzzling.

An absolute normalization uncertainty of 3 - 4% is reasonable for the techniques employed by Laurec et al. While in aggregate their uncertainty estimates are fairly typical, there are a few inconsistencies and problems associated with the error analysis given in the report. For example, the uncertainty associated with determination of the total number of fissions would be expected to be closer to 1.5% (see, e.g., ref. 10). Very little information is given on the flux correction applied to account for any difference in the neutron flux incident on the fission chamber and the target. Further, their approach to combining uncertainties appears to be non-standard. Overall, it is not at all certain why the yields from the ^{239}Pu fission spectrum measurements differ so significantly from other measurements. We conclude that the Laurec results should be taken seriously in any data evaluation and further scrutinized for a more complete understanding of the likely source or sources of the discrepancy with other results.

As an aside, we also note that Nethaway[11] wrote a memo on LLNL results of the 1983 and 1984 fission product measurements in the irradiation of oralloy (93.5% ^{235}U), ^{238}U and ^{239}Pu (93.6% ^{239}Pu, 5.8% ^{240}Pu) with fission spectrum neutrons in the LANL FLATTOP critical assembly and with 14.8-MeV neutrons from the Livermore RTNS-2 facility. We would have liked to consider these results in our analysis but could not due to a lack of detailed documentation. The precision of these measurements, excluding

uncertainties in the number of fissions in a sample, nuclear parameters, etc., is ~ 0.5% for individual nuclides of interest and hence activity ratios will have precisions of ~ 0.7% with many of the systematic errors eliminated. For the FLATTOP measurements, Nethaway used an average of the cumulative yields of ^{95}Zr, ^{99}Mo, ^{140}Ba, ^{144}Ce, and ^{147}Nd given in the LLNL compilation UCRL-51458 (1973) for the determination of the number of fissions in a sample. If current evaluations lead to significant changes in the yields of these isotopes, the cumulative yields reported in the Nethaway memo should be updated. We note that the ^{144}Ce chain yield estimated in the fission spectrum irradiation is considerably higher than that given in the LLNL compilation. We also note that in one irradiation, both a ^{235}U and ^{239}Pu target were included. This raises the possibility of calculating ratios of chain yields in ^{239}Pu to the better-known chain yields in ^{235}U if differences in the neutron flux between the two targets can be estimated accurately. Although most experimental details are not included in the memo, the original files on the experiments still exist. Because it is unlikely that new measurements will be made at the FLATTOP critical assembly, this experiment should be re-examined in detail with a view toward improving the accuracy in the total number of fissions produced and the measured cumulative yields, and a detailed report prepared for peer review.

IV. Energy Dependence of Fission Yields

1. General

Essentially all experimental data indicate that the energy dependence of fission yields in the immediate vicinity of the light- and heavy-massed peaks in the fission of ^{235}U and ^{239}Pu for neutron energies $E_n \leq$ 4-6 MeV must be small. It is also well known that chain yields in the vicinity of symmetric fission demonstrate monotonic increases in the same energy range and that yields in wings of the mass distribution are energy dependent. The principal issues are when and by what magnitude energy dependence affects fission yields in the mass range that includes the interesting nuclides with A ≥ 144 and A ~ 95-99.

Because of its key importance and the difference between the yields used by LLNL and LANL, the cumulative yield of ^{147}Nd has been considered of central interest. The yields used by the two laboratories for irradiations in a "fast" neutron spectrum are about 1.97% and 2.07%, respectively, and differ by a factor of about 1.05. Because the uncertainties in the most reliable individual yield measurements are about $1\sigma =$ ~ 4 - 5%, considerable effort has been expended in applying simple and sophisticated statistical analyses to combine results from many measurements to reduce the uncertainty in the cumulative yield. While certainly very useful, such approaches will always be subject to question because of the possible presence of systematic uncertainties.

At the present time, we know of no theoretical basis for predicting the magnitudes and functional dependence of cumulative yields on energy. In the absence of such it is necessary to examine experimental measurements with sufficiently small absolute

uncertainties that the energy dependence can be established with certainty over the range in neutron energy and mass of interest to the present problem. For the particular case of the cumulative yield of ^{147}Nd, all current evidence suggests that the uncertainties in individual absolute yield determinations obtained with radiochemical methodology are comparable to or larger than the energy dependence that might exist.

The experimental evidence for the energy dependence of fission yields was addressed in some detail by Maeck et al. in two reports.[4,12] Using the cumulative yield ratio ^{150}Nd / ^{143}Nd as an energy index and restricting attention to fission yields derived from isotope dilution methodology, Maeck demonstrated that energy dependence of many cumulative fission yields was suggested over the effective average neutron energy range of about 0 - 1.3 MeV. These reports contain graphs representing both the isotopic abundance of a nuclide and the absolute cumulative fission yield of a nuclide as a function of ^{150}Nd / ^{143}Nd for all fission products studied at that laboratory. While the correlations presented are somewhat crude, Maeck's treatment is both interesting and highly informative.

The relative isotopic abundances of elements produced in fission are one of the primary products of isotope dilution methodology. Such data are unique in the sense that they represent a measure of fission product yields with the minimum error of any set of data thus far produced. Fortunately, the isotopic abundances of fission product Nd and Sm span the mass range $143 \leq A \leq 154$ where Maeck's correlations and the much more global correlation of Wahl[13] both suggest significant energy dependencies and a transition from negative to positive energy gradients.

We have attempted to use all relative isotopic abundances and absolute cumulative yields from isotopic dilution experiments to search for energy dependencies in this mass region as well as the mass region A ~ 95 - 100. The basis for this approach is summarized as follows. We consider the ideal case of a single fission product element with i isotopes, all of which represent the last member of their respective mass chains. That is, the cumulative yield of each isotope is the chain yield for that mass number. If CY(A,E) is the chain yield for mass number A and neutron kinetic energy E, the sum of the chain yields of all i isotopes is the total elemental fission yield, $Y_{El}(E)$, i.e.,

$$Y_{El}(E) = \sum_i CY(A_i, E) \qquad (1)$$

The isotopic abundance, $a(A_i,E)$, is the fraction of the total elemental yield represented by the ith isotope and is given by

$$a(A_i, E) = \frac{CY(A_i, E)}{Y_{El}(E)} \qquad (2)$$

The sum of the abundances is unity. The ratio of the chain yield of the ith isotope at the neutron energy E_1 to the chain yield at energy E_2 is given by

$$\frac{CY(A_i,E_1)}{CY(A_i,E_2)} = \frac{Y_{El}(E_1)}{Y_{El}(E_2)} \frac{a(A_i,E_1)}{a(A_i,E_2)} \tag{3}$$

This equation suggests an approach to search for the energy dependence of a chain yield. In principle, this approach has the minimum experimental uncertainty of any method of which we are aware because the systematic errors in the ratios on the right-hand side of equation (3) can be quite small, particularly the ratio of isotopic abundances. We consider three cases. For each we assume that the energy range of interest is so small that wholesale changes in the gross shape of the mass yield distribution function are not possible.

Case 1: No any energy dependence in any chain yield.

If $CY(A,E_1) = CY(A,E_2)$, for all mass numbers, there is no energy dependence of any chain yield. The mass yield distribution function is independent of energy, an elemental yield cannot change and the relative abundances of the isotopes at the two energies must be identical. That is,

$$\frac{a(A_i,E_1)}{a(A_i,E_2)} = 1 \tag{4}$$

Case 2: No energy dependence in the chain yields of interest, but some energy dependence for other mass chains.

If $CY(A,E_1) \neq CY(A,E_2)$ everywhere but over the range of mass numbers of the isotopes we consider $CY(A_i,E_1) = CY(A_i,E_2)$, the elemental yields at the two energies are not the same. They are in the ratio $Y_{El}(E_2) / Y_{El}(E_1) = \text{constant} \neq 1$. That is

$$\frac{a(A_i,E_1)}{a(A_i,E_2)} = \frac{Y_{El}(E_2)}{Y_{El}(E_1)} \neq 1 \tag{5}$$

This could happen, at least approximately, in the following manner. If the neutron energy is increased, chain yields near symmetric fission and in the wings of the distribution increase, and chain yields in other regions must decrease. It is possible that the loss in fission yields is spread over many mass chains throughout the region near the peak maxima and thus they appear to be essentially constant.

Case 3: Energy dependence of the mass chains of interest is expected except for the special case of an accidental cancellation.

If, over the mass range of interest, $CY(A_i,E_1) \neq CY(A_i,E_2)$, then $a_i(E_1) \neq a_i(E_2)$ and there must be an energy dependence of the chain yield except if

$$\frac{Y_{El}(E_2)}{Y_{El}(E_1)} = \frac{CY(A_i, E_2)}{CY(A_i, E_1)} \qquad (6)$$

The latter, which requires that the total elemental yield vary in exactly the same manner as the chain yield, would seem rather improbable. However, it could appear to be true in practice when changes in the mass yield function are small and uncertainties in the experimental data are sufficiently large. Hence the need for data with the minimum possible uncertainties to identify mass yield variations as small as ~ 2%.

Apart from the special case of equation (5), where the isotopic abundance variation is inversely proportional to the variation in elemental yield, the observation that $a_i(E_1) \neq a_i(E_2)$ *means that there must be an energy dependence of the chain yield even if it is not immediately apparent in the measured absolute chain yields because of their larger uncertainties*. Further, because the sum of the isotopic abundances of all i isotopes is unity at each energy, if one of the abundances increases with energy at least one of the others must decrease with energy.

2. The Nuclides and Mass Regions Considered in This Study

In Figure 1 is shown the region of the nuclide chart that includes the Nd and Sm isotopes considered here. The isotopes$^{143\text{-}146,148,150}$Nd are all stable end products of the β-decay chains produced in fission and their fission yields were directly measured in the isotope dilution experiments. ^{142}Nd is a shielded nuclide and the end product of the A = 142 mass chain is ^{142}Ce. Because of their short half-lives, the cumulative yields of ^{147}Nd and ^{149}Nd were not measured. However, these nuclides are sufficiently close to ^{147}Sm and ^{149}Sm, the stable end products of their respective mass chains, that the cumulative yields of the latter can be taken to represent the yields of ^{147}Nd and ^{149}Nd to a good approximation.

Sm144 0+ 3.1	Sm145 340 d 7/2- EC	Sm146 1.03E8 y 0+ α	Sm147 1.06E+11 y 7/2- α 15.0	Sm148 7E+15 y 0+ α 11.3	Sm149 2E+15 y 7/2- 13.8	Sm150 0+ 7.4	Sm151 90 y 5/2- β-	Sm152 0+ 26.7	Sm153 46.284 h 3/2+ *	Sm154 0+ 22.7
Pm143 265 d 5/2+ EC	Pm144 363 d 5- EC	Pm145 17.7 y 5/2+ EC,α	Pm146 5.53 y 3- EC,β-	Pm147 2.6234 y 7/2+ β-	Pm148 5.370 d 1- β- *	Pm149 53.08 h 7/2+ β-	Pm150 2.68 h (1-) β-	Pm151 28.40 h 5/2+ β-	Pm152 4.12 m 1+ β- *	Pm153 5.25 m 5/2- β-
Nd142 0+ 27.13	Nd143 7/2- 12.18	Nd144 2.29E+15 y 0+ α 23.80	Nd145 7/2- 8.30	Nd146 0+ 17.19	Nd147 10.98 d 5/2- β-	Nd148 0+ 5.76	Nd149 1.728 h 5/2- β-	Nd150 1.1E19 y 0+ β- 5.64	Nd151 12.44 m 3/2+ β-	Nd152 11.4 m 0+ β-

Figure 1. The region of the nuclide chart in the vicinity of the Nd and Sm nuclides considered here.

The yields of ^{147,149}Sm as well as those of mass numbers A = 151, 152 and 154 were directly measured or inferred after application of necessary corrections in most of the isotope dilution experiments. Because of the long half-life of ^{147}Pm and the relatively short time allowed for decay after the end of irradiation before chemical processing of a target began, a significant correction for undecayed ^{147}Pm was required to extract the

^{147}Sm cumulative yield. The cumulative yields of the majority of the Sm isotopes in thermal irradiations required a large number of corrections because of the large capture cross sections of these isotopes and thus are subject to some question. A small decay correction was generally necessary to obtain the cumulative yield of ^{151}Sm.

In Figure 2 is shown the region of the nuclide chart containing the stable and long-lived isotopes of Zr and Mo. The cumulative yields of the Zr isotopes of A = 91-94 and 96 were directly measured, the yield for the A = 90 mass chain being represented by the cumulative yield of ^{90}Sr. Cumulative yields were also directly measured for the Mo isotopes A = 95, 97, 98 and 100. The cumulative yield for ^{99}Mo was not measured because of its short half-life. This cumulative yield of ^{99}Tc, which would represent the ^{99}Mo yield, could not be measured with the isotope dilution technique.

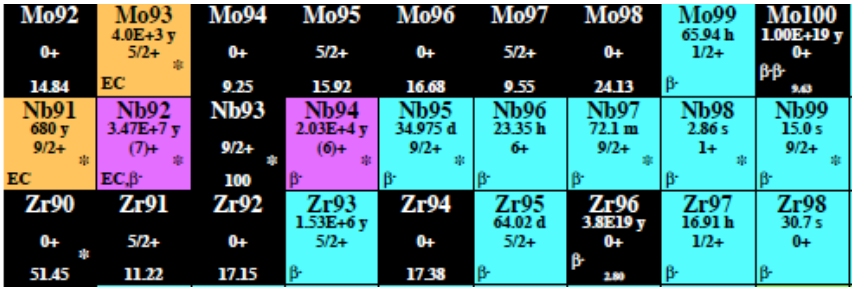

Figure 2. The region of nuclide chart in the vicinity of the Zr and Mo nuclides considered here.

3. Relative Isotopic Abundances of Fission Product Nd

The Nd isotopes of mass numbers A = 143-146, 148 and 150 are either the stable end products of mass chains or sufficiently near the ends of these chains that their cumulative yields can be taken to be the chain yields. The pooled fast neutron evaluations of England and Rider[14] support this conclusion with the lone possible exception of A = 150. The evaluation indicates that about 0.22% of the A = 150 chain yield lies beyond ^{150}Nd and thus small changes in the mean charge of the distribution of independent fission yields, Z_p, or changes in the width of the distribution at elevated neutron energies might affect the interpretation of the cumulative yield of ^{150}Nd. With this qualification, the Nd isotopes fulfill the requirements of chain yield monitors.

There are a total of nine[a] essentially independent isotope dilution experiments described in refs. 2, 3, and 5-8. The absolute elemental yields of Nd were reported for eight of the experiments and these are given in Table 1 along with the estimated average neutron energies in each experiment and the estimated uncertainties (± 1σ) in the parameters. The neutron energies were estimated with the correlation of Figure 1 of ref. 4. The average

[a] The absolute chain yields reported by Gabeskiriya et al.[7] are tied to the sum of the yields for ^{145,146}Nd given by Lisman et al.[5] but the isotopic abundances are not.

elemental yield of Nd over the energy range $0.21 \leq < E_n > \leq 1.33$ MeV is found to be $16.16 \pm 0.19\%$. A simple linear fit to the data returns a slope whose uncertainty is about a factor of 100 smaller. With ~ 95% confidence, there is less than a 2.4% variation in the elemental yield of Nd at the $\pm 2\sigma$ limit and any systematic energy dependence must be significantly smaller. Conservatively, the observation of changes in the relative abundances of the Nd isotopes as a function of neutron energy that are significantly larger than about ± 2.4% must be indicative of changes in the shape of mass yield distribution function and therefore indicative of an energy dependence of the chain yields over the mass and energy ranges considered.

Table 1. Absolute elemental yields for Nd from isotope dilution experiments in fast reactors. The abbreviation CAPS stands for capsule and represents one of the 4 fueled capsules irradiated in EBR-II Row 8. Because of their different locations, each of the capsules was subjected to a neutron flux distribution of different shape.

Experiment	$< E_n >$ (keV)	$\sigma (E_n)$ (keV)	$Y_{El}(E)$ Nd (%)	$\sigma (Y_{El}(E))$ (%)
EBR-II CAPS 35	210	15	15.924	0.077
EBR-II CAPS 26	440	9	16.101	0.080
EBR-II CAPS 5	450	24	16.163	0.078
EBR-II CAPS 24	480	13	16.369	0.077
TACO RAPSODIE	594	160	15.87	0.46
DOUNREAY	720	180	16.18000	0.191
BOR-60	735	55	16.37000	0.080
EBR-I	1330	70	16.33000	0.129
$< Y_{El}(E) >$			16.16 ± 0.19	

Relative isotopic abundances were directly reported for all of the experiments described in refs. 1-3, and 5-7. These experiments were performed within the same laboratory and by at least a core of the same personnel. As such they appear to represent a data set for which random and systematic errors are very likely relatively constant or were reduced as a function of time and experience. The reports cited provide great detail on the experiments, methodologies, error assessment and analysis. We will refer to these collectively as the Maeck experiments. For some of the remaining experiments, described in refs. 6 and 8, only absolute fission yields were reported. These were converted to relative isotopic yields but, of necessity, are assigned uncertainties associated with the absolute yield estimates.

To search for possible energy dependences in a simple manner, the isotopic abundances at each energy were divided by the corresponding isotopic abundances for thermal neutron fission as reported by Maeck et al.[1,] The resultant data, that represent the ratios $a(A_i,E_2)/a(A_i,E_1)$ of equation (3), are shown in Figure 3 as a function of the estimated mean neutron energies given in Table 1. The curves in the figure represent simple least

squares fits to the data for each mass number with a quadratic function forced to go to unity at $< E_n > = 0$ [b] and for which no errors were included in the fits. These curves have no real physical meaning and were chosen only to provide good representations of the trends in the data. The dashed horizontal line represents the case of no energy dependence and the horizontal dotted lines represent the ± 2.4% uncertainty at the 2σ limit of the average elemental abundance discussed above.

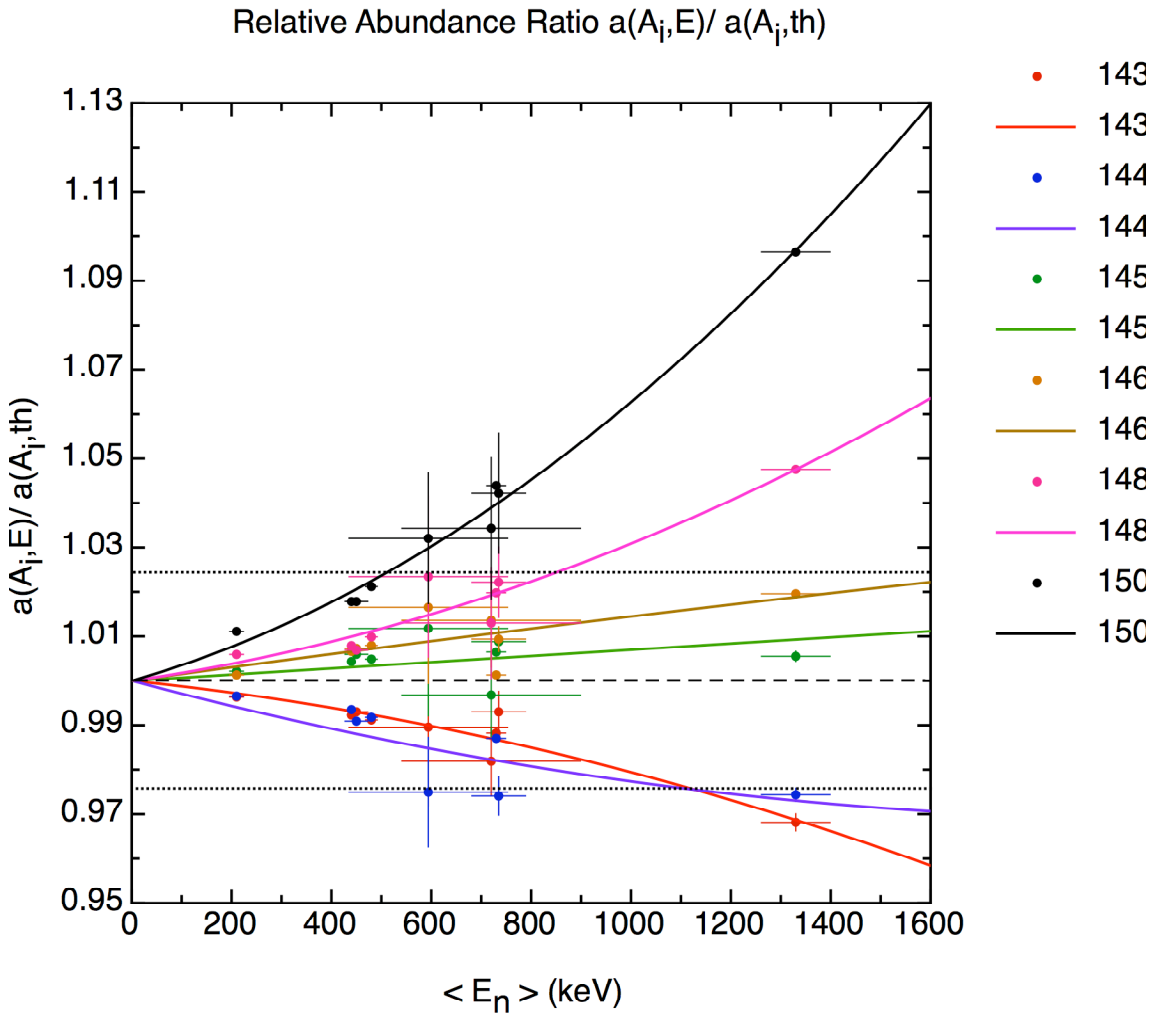

Figure 3. Isotopic abundances of the Nd isotopes as a function of average neutron energy divided by the thermal abundances of Maeck et al.[1]

The data in Figure 3 are quite remarkable because of their internal consistency and the systematics they suggest. The majority of the data shows very little scatter about a smooth energy variation. With the exception of the relative isotopic abundances for A =

[b] This procedure may be questioned because of the spin of the ground state of ^{239}Pu. See Section VI.

143 and A = 144, the data display a regular increase in slope with increase in mass number. The data suggest that for average neutron energies greater than about 1 MeV, the chain yields for A ≥ 146 are likely to be greater than those found at thermal energies while for A ≤ 144 the chain yields are likely less than those found at thermal energies. The data also suggest that the energy dependence of the chain yield, at least in this region for this fissioning system, is not necessarily linear. We stress this point because both Maeck[4, 12] and the LANL statistical analyses[15] we have seen tacitly assume a linear variation.

The conclusions given above are subject to uncertainty in a number of respects. They rely on a limited data set for which the mass chains A = 147 and A = 149 are missing. This is of some concern because of the possible presence of an odd-even variation in the energy dependence of the chain yields. The suggestion of non-linearity in the energy dependence of at least some chain yields is strongly influenced by the data for $< E_n > \sim 1330$ keV from the experiment by Lisman et al. in EBR-I.[5] All of the experiments considered were performed in fast reactor irradiations, and because of the limited range in average neutron energy probed, projections of isotopic energy variations to an average energy of ~ 1.9 MeV that is characteristic of the critical assembly Jezebel cannot be made with any certainty.

The average neutron energy associated with an experiment considered here has uncertainty from two sources. It depends on the quality of the curve drawn in Figure 1 of ref. 4 and whether it represents the average neutron energy in the spectrum or the average neutron energy producing fission. (Both questions may be removed by access to reports not available to us at the time of this writing.) More importantly, we do not have access to the details of the experimental measurements used to define the neutron distributions at the various irradiation sites in the fast reactors. In some cases it appears that the reported neutron energies may be those from reactor physics calculations that are also unavailable to us.

Finally we note the absence of data from two reports by Robin et al.[16,17] that were referenced by Maeck in his reports. The experiments were performed in the Rapsodie reactor at estimated average neutron energies of roughly 730 keV and 1200 keV. We do not have the references containing the Robin reports and Maeck does not list the data and their uncertainties. We have taken estimates of the isotopic abundances for the higher-energy measurement from Maeck's graphs and they are reasonably consistent with the trend in the data shown in Figure 3 with the exception of the trends for A = 145 and A = 146. Assuming uncertainties comparable to those of the Maeck data, the trend line for the energy dependence for A = 145 would tend to be slightly negative and that for A = 146 slightly less positive.

4. The Sm Isotopes from Isotope Dilution Experiments

The isotopic abundances of Sm fission products with mass numbers A = 147, 149, 151, 152 and 154 were measured only in the experiments described in ref. 1-3, 5 and 8 with one exception (see below). A significant problem with assessing the quality of the

thermal data arises from the rather large corrections required for neutron capture in a number of isotopes. In particular, Maeck[1] notes that a large number of corrections were required for the thermal data, affecting the comparison between fast and thermal data. He also notes that there is some question concerning the abundance of ^{154}Sm. Our review of the corrections applied by Maeck indicates that they are correct in principle.

The total elemental yield for the Sm isotopes as a function of average neutron energy from the seven experiments reported in the literature are shown in Figure 4 along with linear and quadratic fits to the data. The data imply an increase in fission yield by a factor of about 1.12 over the energy range thermal - 1.33 MeV.

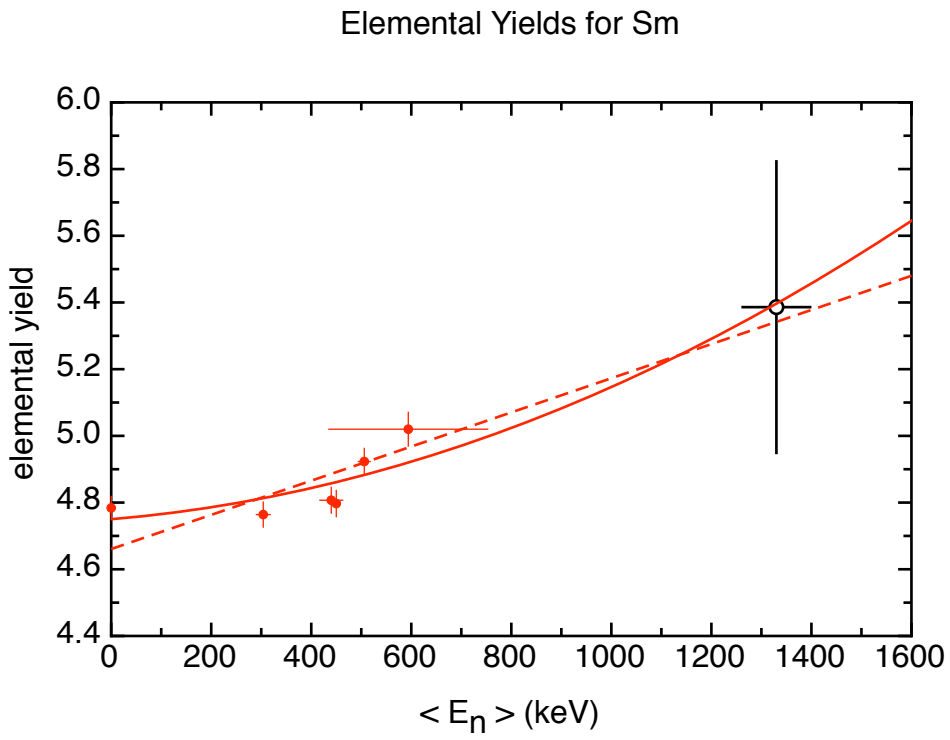

Figure 4. Elemental fission yields of the Sm Isotopes. The data point shown in black was obtained with an estimate of the cumulative yield of ^{147}Sm. The large uncertainty assigned to this estimate dominates the total elemental yield shown in the figure.

The datum at 1.33 MeV is due to Lisman et al.[5] from the experiment in EBR-I. The isotopic abundances of all Sm isotopes noted above were measured with the exception of that for ^{147}Sm. The yield for the later was estimated by linear interpolation between measured cumulative yields and was assigned an arbitrary uncertainty of ± 20% and this uncertainty dominates the total uncertainty in the elemental yield reported by Lisman et al. While the assignment of such a large uncertainty seems overly conservative, we retain it as given by the authors. Dismissing the Lisman et al. datum, the data below 600 keV suggest an average slope of roughly 0.4% MeV^{-1} (absolute) for the elemental abundance.

The ratios of the isotopic abundances at energy $< E_n >$ to the Maeck et al.[1] isotopic abundances at thermal energies are shown in Figure 5 along with linear fits for each mass number. The fits were performed without the requirement that they go to unity at thermal energies. The data and fits demonstrate the discrepancy noted by Maeck concerning the consistency between the thermal isotopic abundances and those from the fast neutron irradiations. The intercepts at $< E_n > = 0$ keV deviate from unity by as much as 5-7%. Whether these reflect uncertainties in the corrections applied by Maeck or represent real physical effects is not known. Nevertheless, the simple linear fits suggest a trend in the data related to mass number: the isotopic abundances for $A \leq 151$ decrease with energy while those for A = 152 and 154 increase with increasing energy. We note that the datum from the TACO experiment[8] at $< E_n > \sim 600$ keV is somewhat discrepant from those of Maeck. However, it does not change the general nature of the trend in the data.

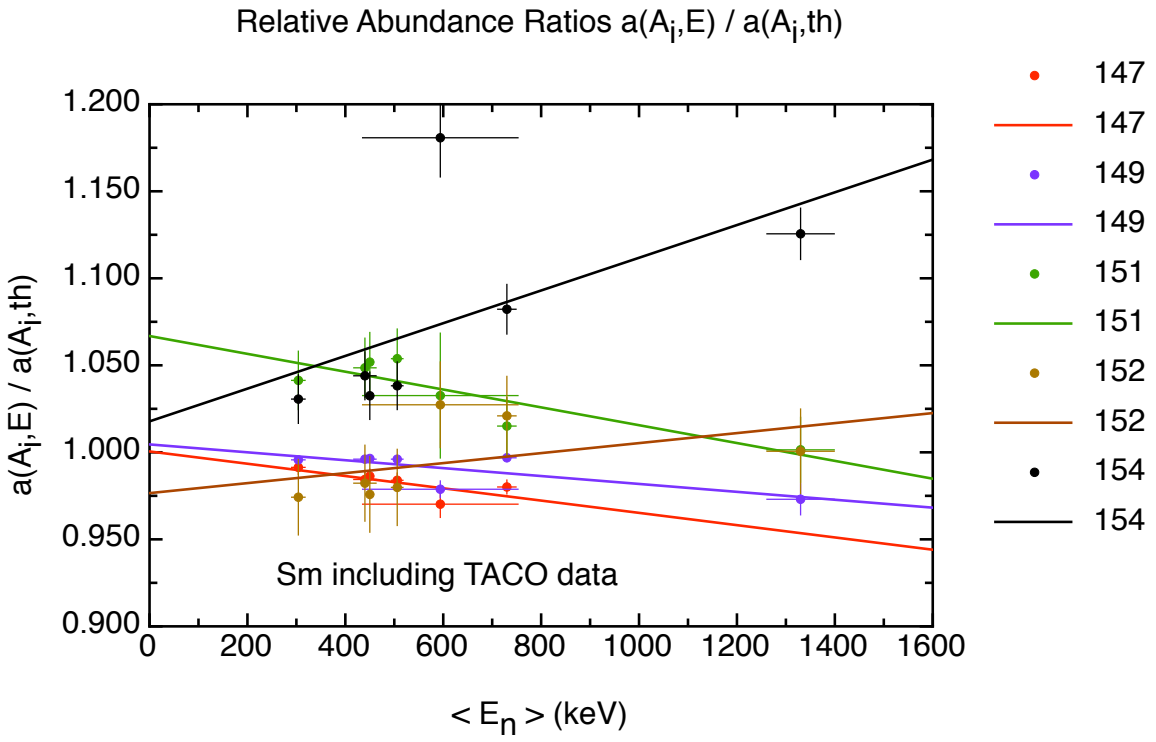

Figure 5. Ratios of isotopic abundances of the Sm isotopes to the isotopic abundances at thermal energies.[1]

5. Cumulative Fission Yields for the Nd and Sm Isotopes

The cumulative fission yields of the Nd and Sm isotopes cover the entire mass range of interest for examining the possible energy dependencies of the nuclides ^{144}Ce and ^{147}Nd. We have used the cumulative yields from all of the experiments where they have been reported to prepare Figures 6 and 7 that cover the mass ranges $143 \leq A \leq 147$ and $146 \leq$

A ≤ 150, respectively. Again, the curves shown in the figures are quadratic fits to the data meant solely for the purpose of correlations. Also, the datum from the Lisman et al.[5] experiment for A = 147 that was obtained by linear interpolation between known cumulative yields is not shown.

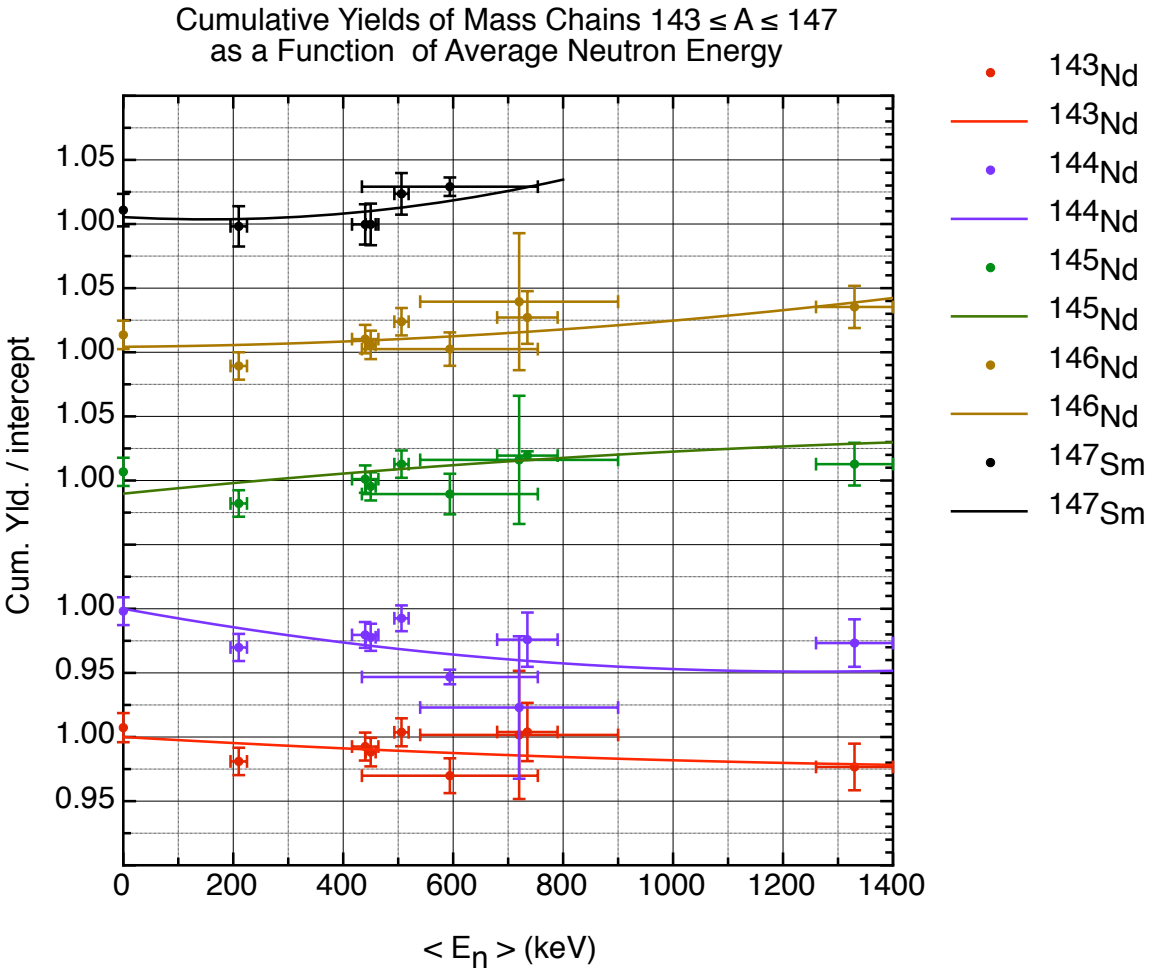

Figure 6. Cumulative yields divided by fit intercept for Nd and Sm isotopes with mass numbers 143 ≤ A ≤ 147 versus average neutron energy in an experiment.

Notwithstanding the increased uncertainties associated with conversion of isotopic abundances to absolute cumulative yields, the data in Figure 6 demonstrate a simple systematic trend as a function of energy and mass number. The cumulative yields for A ≤ 144 are consistent with decreasing yields with increasing energy and *vice versa* for the cumulative yields for A ≥ 145. The data in Figure 7 are consistent with these trends. They imply consistency between the yield measurements for the Nd and Sm isotopes and also suggest that the energy dependence of the cumulative yields generally increases with mass number in this region. Although not shown, this trend continues in the data for A =

152 and A = 154. The limited data for A = 147 combined with this trend support an increase in the cumulative yield with increasing average neutron energy.

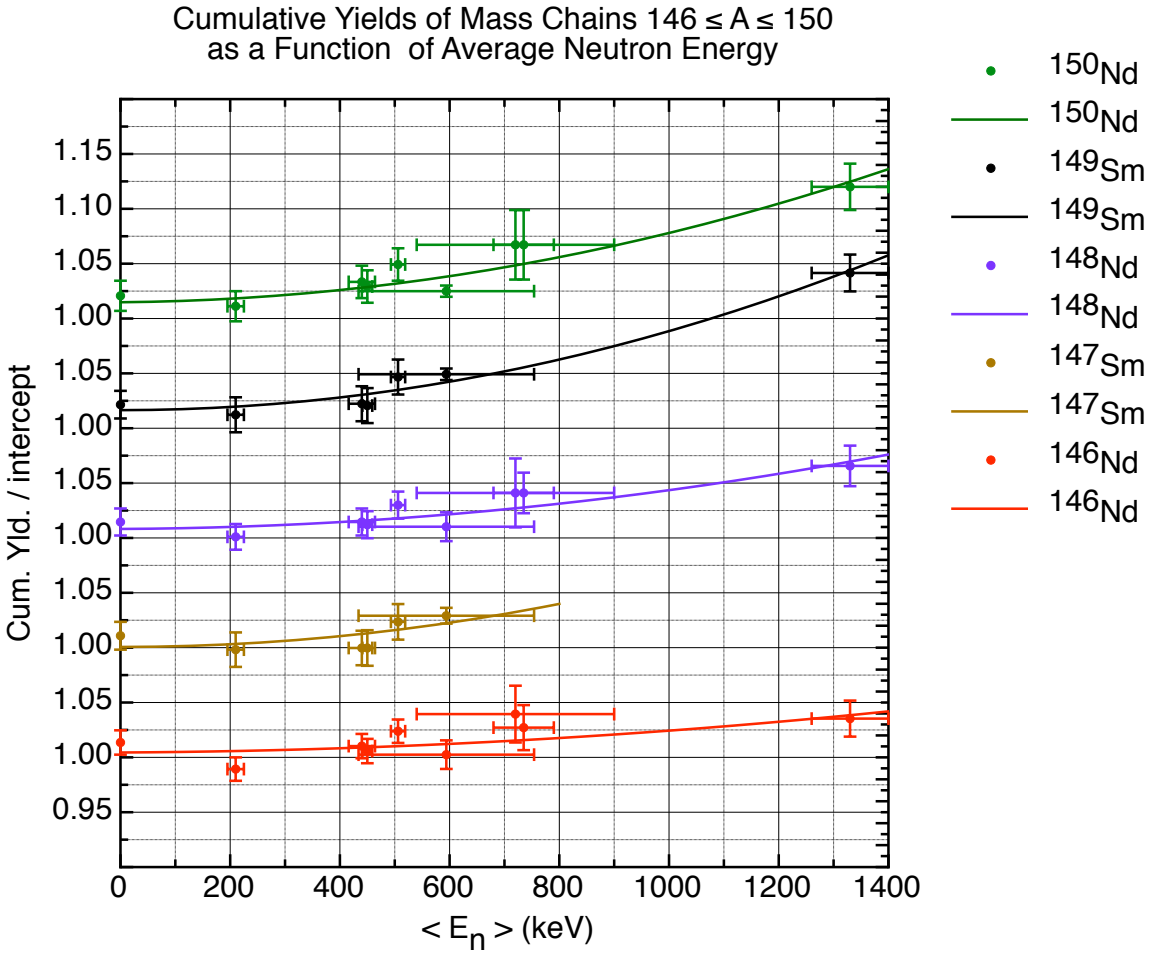

Figure 7. Cumulative yields divided by fit intercept for Nd and Sm isotopes with mass numbers 146 ≤ A ≤ 150 versus average neutron energy in an experiment.

6. Zr from the Isotope Dilution Experiments

In the light-massed peak from n + ^{239}Pu fission, zirconium possesses five isotopes that represent the chain yields for A = 91 - 94 and 96. The relative isotopic abundances were measured in seven experiments, including thermal fission, but the absolute abundances were reported for only six of these. Because of dissolution problems, the experiment in EBR-II Row 4 [3] at $< E_n > \sim 0.73$ MeV only provided isotopic abundances. All of the data were produced by the Maeck group. Maeck notes that the experiments involving fission product Zr represented one of the more difficult analytical challenges they faced. They found very significant contamination from natural Zr, presumably from the quartz containment of the dissolution apparatus. For the experiments at thermal energies, that

involved burnups of about 1 atom percent, Maeck indicated that as much as a 70% correction was required for the presence of natural contamination. As a result, isotopic abundances were only reported for the high-burnup (~ 50 atom percent) experiments.

The elemental yields reported for Zr are given in Table 2 and the data are shown versus average neutron energy in Figure 8.

Table 2. Elemental yields for Zr from the isotope dilution experiments of Maeck et al.

$< E_n >$ (MeV)	$\sigma < E_n >$	Elemental Yield (%)	σ (El. Yld.) (%)
0	0	18.125	0.126
210	15	18.287	0.148
440	24	18.254	0.144
450	9	18.322	0.226
506	13	18.772	0.146
1330	70	19.120	0.165

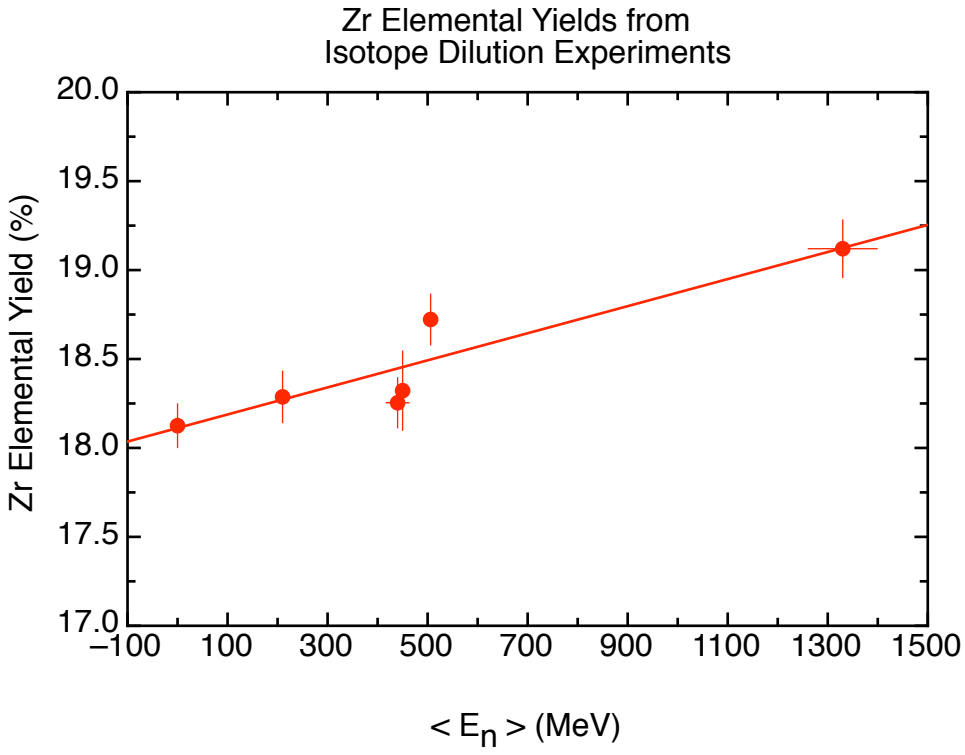

Figure 8. Elemental yields for Zr from the experiments of Maeck.

The data demonstrate an increase in elemental yield by a factor of about 1.05 over the energy range thermal $\leq < E_n > \leq$ 1.33 MeV and the linear fit shown in the figure has a slope larger than its estimated uncertainty by a factor of about 5.

The evaluated data of England and Rider [14] provide thermal and pooled fast neutron total chain yields for the mass chains involved of 18.43% and 18.28%, respectively. These are clearly inconsistent with the Maeck results. We know of no publication that directly discusses the energy dependency of the Zr elemental yield implied by these data.

In Figure 9 are the reported isotopic abundances divided by the isotopic abundances for thermal fission from Maeck for the seven experiments for which such data were reported along with linear fits for each mass number. The scatter in the data is relatively large, especially for the experiment at $< E_n > \sim 730$ keV. Other than being consistent with small energy dependencies, the scatter in the data precludes further conclusions.

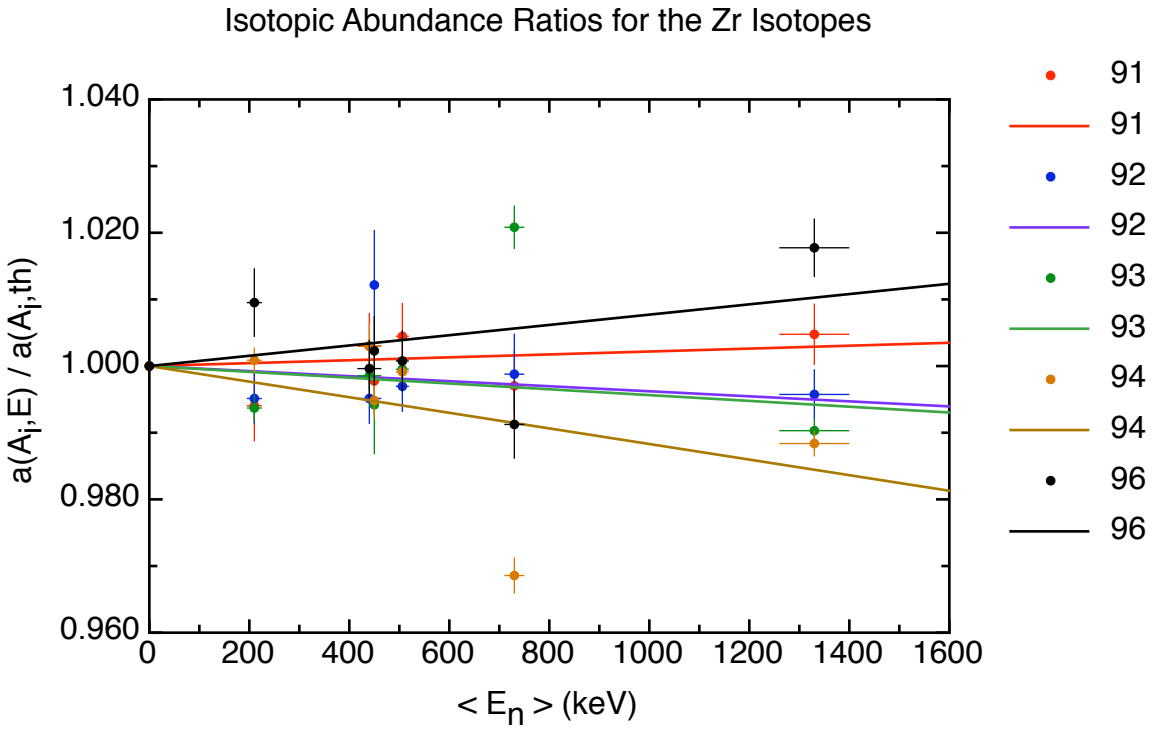

Figure 9. Ratios of isotopic abundances for the Zr isotopes to the isotopic abundances at thermal energies for the Maeck experiments.

In Figure 10 are shown the ratios of the absolute fission yields of the Zr isotopes to the absolute thermal fission yields as a function of average neutron energy. Because all of the ratios have uncertainties of about 2.5%, only those for A = 91 are shown. Within the estimated uncertainties, the data for all mass numbers show very nearly the same energy dependency as expected from the data in Figure 6 and imply that at least some cumulative fission yields for the Zr isotopes might display significant energy dependencies.

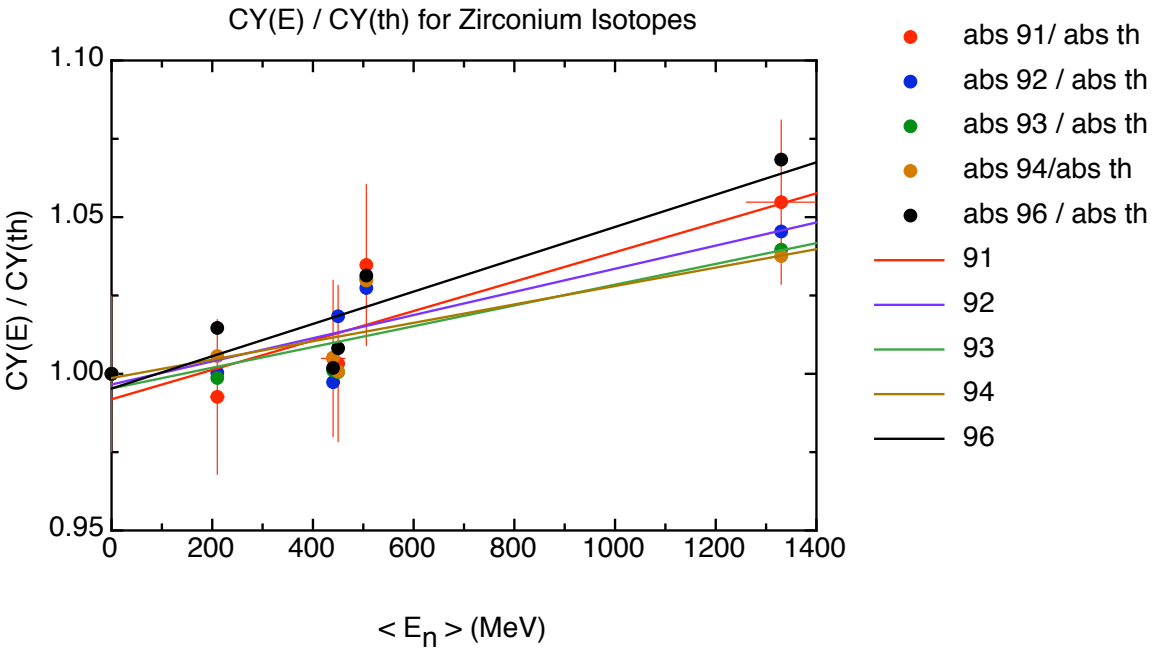

Figure 10. Ratios of absolute abundances of Zr isotopes to the abundances at thermal energies.

7. Mo from the Isotope Dilution Experiments

Mo has four isotopes for which isotopic abundances and absolute fission yields have been reported in isotope dilution experiments. They are restricted to the experiments from the Maeck group. Further, because of incomplete dissolution of small quantities of the target, no isotopic abundances or absolute yields were reported from the thermal measurements. In this experiment, the PuO_2 fuel was mixed with Al powder and metallic inclusions, long known from the behavior of oxide reactor fuels, formed rapidly. This target composition was not used in any other experiments.

The elemental yields from the five experiments reporting them are given in Table 3. The average elemental yield is 22.58 ± 0.23%, corresponding to a 1σ relative error of ± 1.0 %. The elemental yields are shown in Figure 11 as a function of the average neutron energies along with a linear fit to the data. The slope of the fit is zero within its estimated uncertainty. For reference, the ratio of the predicted yield at 1.9 MeV to that at thermal energies calculated from the linear fit is 1.015 ± 0.021.

Table 3. Elemental Mo yields from experiments conducted by the Maeck group.

$< E_n >$ (MeV)	$\sigma < E_n >$	Elemental Yield (%)	σ (El. Yld.) (%)
0	0	-	-
210	15	22.726	0.145
440	24	22.443	0.148
450	9	22.255	0.142
480	13	22.670	0.143
1330	70	22.820	0.213
$< Y_{El}(E) >$		22.582	0.230

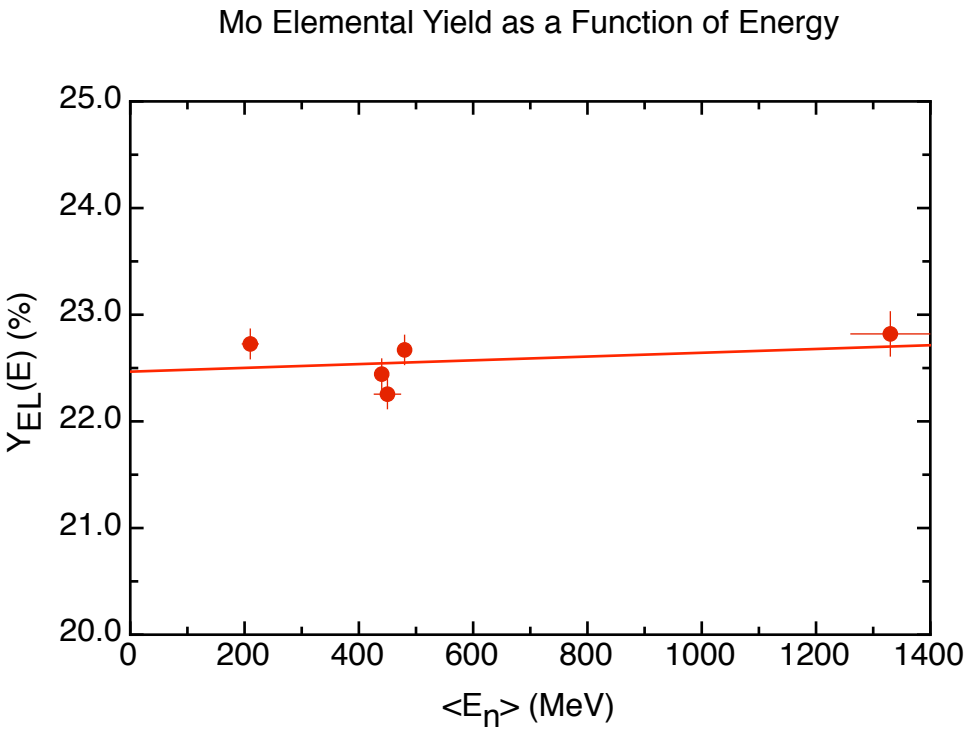

Figure 11. Elemental yields for Mo from the experimental data of Maeck.

In the absence of isotopic abundances at thermal energies, we have simply plotted the relative isotopic data at elevated neutron energies along with unweighted linear fits with zero slopes and with quadratic fits. These are shown in Figure 12. The constant fits to the data seem rather good and the fitting parameters they provide are given in Table 4.

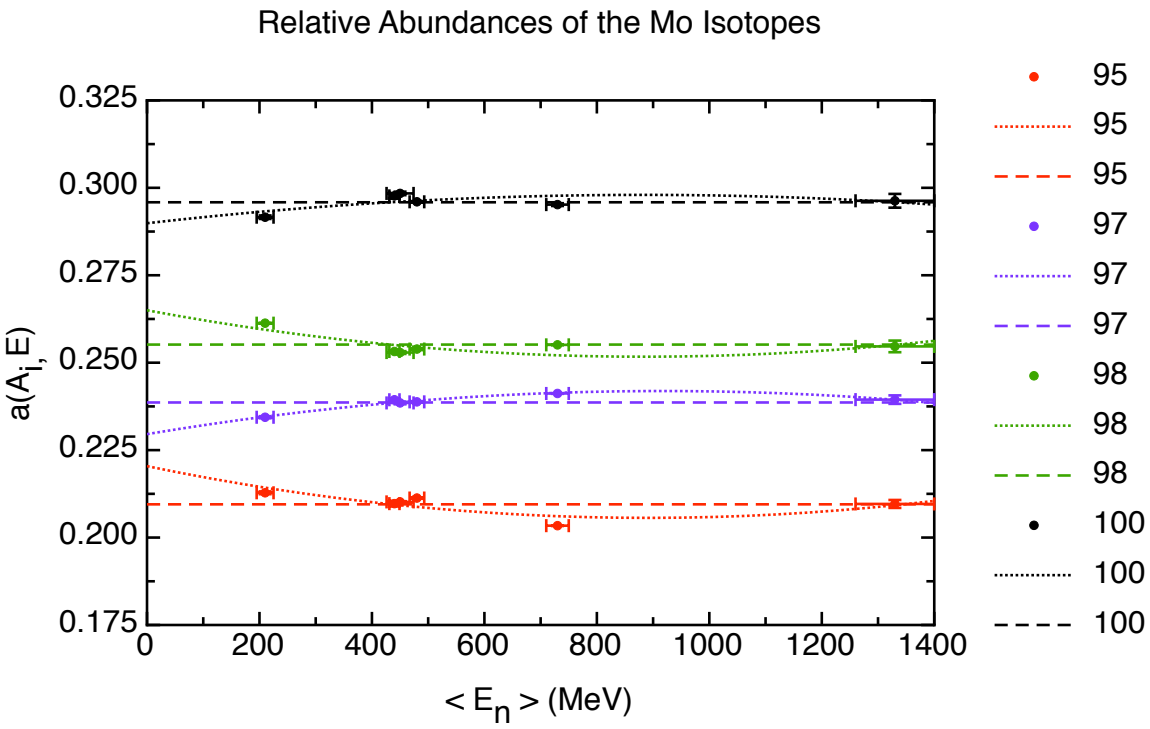

Figure 12. Isotopic abundances of the Mo isotopes from the Maeck data.

Table 4. Fitting parameters from the isotopic abundance data from the Maeck group.

A	Constant	σ (Constant)	σ / Constant
95	0.2095	3.22×10^{-3}	1.54×10^{-2}
97	0.2386	2.27×10^{-3}	9.51×10^{-3}
98	0.2552	3.11×10^{-3}	1.22×10^{-2}
100	0.2959	2.39×10^{-3}	8.08×10^{-3}

The largest uncertainty in a constant is ± 1.54% (relative) for ^{95}Mo. The sum of the constants is 0.9992 ± 0.0056, in agreement with the assumption that the isotopic yields are independent of energy. Coupled with the data on elemental yield, the data strongly imply that the fission yields of the Mo isotopes are independent of energy within 1-2% for average neutron energies of $0.2 \leq \langle E_n \rangle \leq 1.3$ MeV.

V. Comparison with Radiochemical Data

1. ^{147}Nd Data from Isotope Dilution and Critical Assembly Experiments

The LANL and LLNL radiochemistry groups have performed a large number of experiments on various critical assemblies over the years. They were all based on radiochemical methodologies. They are certainly of high quality and probably represent

some of the best work with the radiochemical approach. The reported errors in individual measurements are on the order of 4-5% or about twice those reported for individual measurements with isotope dilution methodologies.

Data from the critical assembly experiments performed at LANL were kindly provided by Mark Chadwick[18] and we treat them as received. In Figure 13 are shown the absolute cumulative yields for ^{147}Nd as a function of the average neutron energies producing fission calculated with Monte Carlo simulations. Included in the figure is a linear least squares fit to the data, including estimated uncertainties. The slope of the linear fit was $(5.570 \pm 4.813) \times 10^{-5}$ % keV^{-1} and is consistent with a slope greater than zero at the level of about 1.2 σ. The cumulative yield calculated from this fit at an average neutron energy of 1.5 MeV, the reference energy chosen by LANL, is 2.069 ± 0.097%. The error includes both the estimated error in the slope and the estimated error of the intercept. Within the uncertainty of ± 1σ, the calculated cumulative yield for ^{147}Nd lies within the range 1.97 - 2.17%.

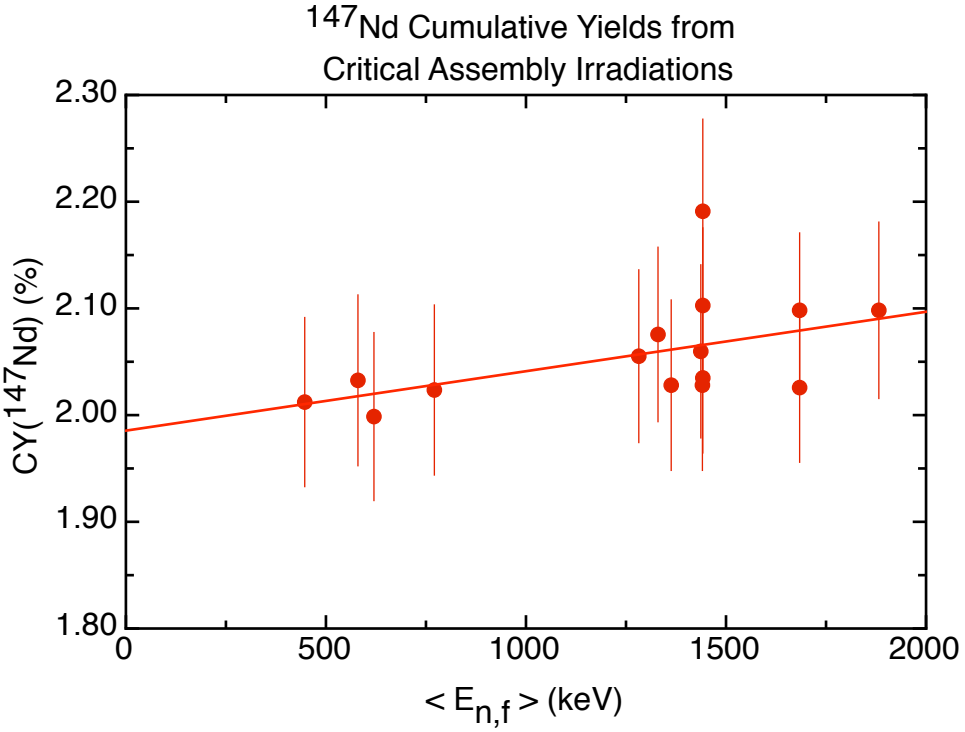

Figure 13. Cumulative yields for ^{147}Nd from critical assembly experiments.[18]

In Figure 14 are shown the data from the same critical assembly measurements along with those from the isotope dilution experiments as provided and interpreted by Chadwick.[18] The linear fit to this data set now provides an estimated yield for ^{147}Nd at $< E_n > = 1.5$ MeV of 2.082 ± 0.041%. Within an uncertainty of ± 1σ, the estimated cumulative yield for ^{147}Nd lies in the range 2.04 - 2.12%.

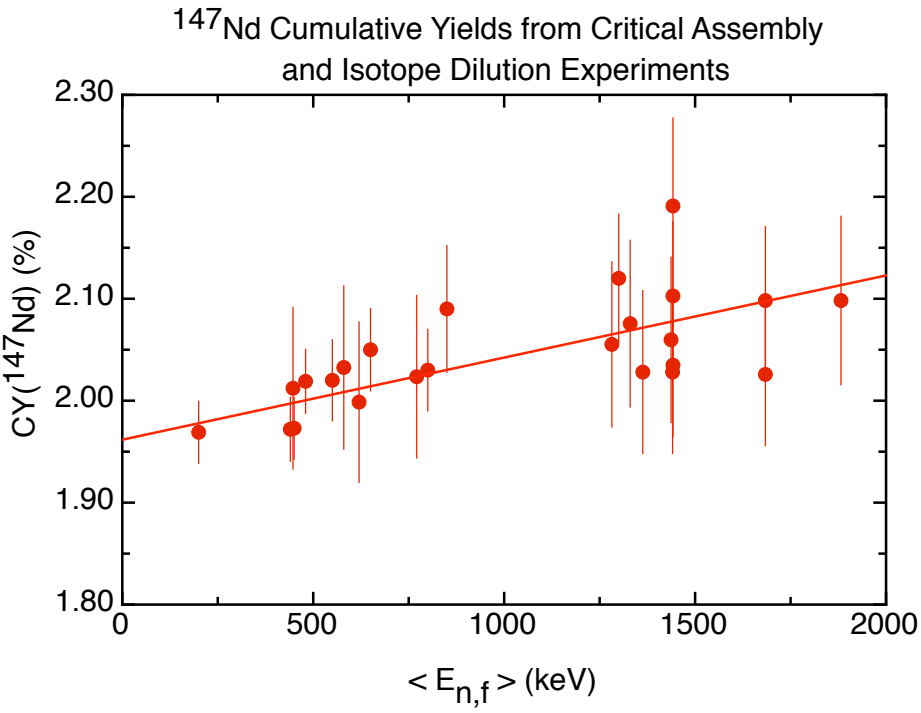

Figure 14. Cumulative fission yields for ^{147}Nd for critical assembly and isotope dilution experiments from ref. 18.

From this simple analysis, it seems clear that it is the data from the isotope dilution experiments that tend to control the suggestion that there is significant energy dependence in the cumulative yield of ^{147}Nd.

As a rough means of placing the yields estimated from the LANL critical assembly measurements on the same footing as those from the relative isotopic abundance measurements, we have divided the cumulative yields from the critical assembly experiments by the same thermal yield from Maeck as used to provide the data in Figure 3. The results are shown in Figure 15 along with the fitted curves from Figure 1 and a dotted-dashed line representing a linear fit through the relative critical assembly data. Such a comparison is reasonable if the elemental yield of Nd is independent of energy. Figure 15 emphasizes the relatively large uncertainties in the radiochemical measurements as well as their relatively large scatter as compared to that found in the isotope dilution experiments. Nevertheless, it seems clear that there is general agreement between the data from the two sets of experiments. It seems equally clear that the extent of agreement between the two data sets with respect to a functional dependence of the ^{147}Nd cumulative yield on average neutron energy is highly uncertain.

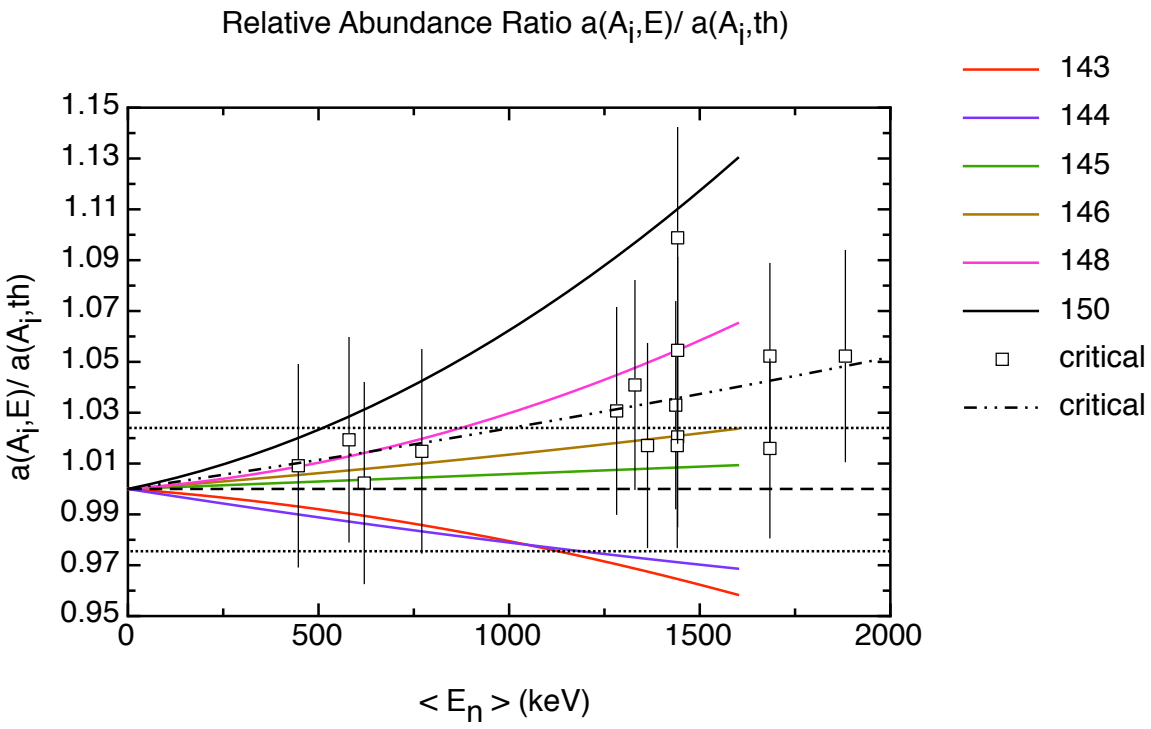

Figure 15. Comparison of the abundance ratios of the Nd isotopes from Figure 1 with the relative cumulative fission yields from the critical assembly experiments[18].

2. Comparison of Fission Yields for Nuclides of Interest

In this section we attempt to compare the best estimates for the cumulative yields of the nuclides of interest with those recommended by LLNL and LANL. This is not as straightforward as it seems because, for the LLNL yields, we do not know exactly to which neutron spectrum the yields for ^{95}Zr, ^{99}Mo, and ^{144}Ce refer. The most current LANL recommendations are for a neutron spectrum with an average energy of ~ 1.5 MeV[18, 19]. The most current recommendations from LLNL are from ref. 20 and the energy dependence of the ^{147}Nd cumulative yield is from ref. 21. In addition, our primary reviews have been concerned only with data from the isotope dilution experiments with a maximum average neutron energy of ~ 1.3 MeV. As a result, the comparisons given below can only be used to give a rough picture of the current state of the cumulative yields of interest. And it is important to remember that all of our estimates do not permit conclusive projections to cumulative yields at an average neutron energy of ~ 1.9 MeV characteristic of the critical assembly Jezebel.

Given these considerations, we have chosen the average neutron energy of 1.5 MeV to present some comparisons and we simply provide the data from the preliminary evaluations of LLNL as they are. Our estimates are based on extrapolations from the maximum energy of ~ 1.3 MeV in the isotope dilution experiments and were performed with the two different assumptions

(a). The fitted quadratic curves shown in Figures 3 and 15 represent good representations of the data from the isotope dilution experiments.
(b). The data from the isotope dilution experiments are best represented by linear variations with average neutron energy.

The errors associated with our projections have been estimated from the errors in the absolute yields used and estimates of errors from the functional fits to the data. Thus they represent the *minimum* uncertainty that can be assigned to the projections. Assumptions specific to the estimated cumulative yields of individual nuclides are included in the footnotes to the table.

In Table 5a we present the current state of fission yield recommendations for the nuclides ^{95}Zr, ^{99}Mo, ^{144}Ce and ^{147}Nd from LANL, those that we project to an average neutron energy of 1.5 MeV from our analysis of the data from the isotope dilution experiments using quadratic (Analysis I) or linear fits (Analysis II) to the data, and the ratios of the LANL recommendations to those from our projections. Similarly, Table 5b presents comparisons with the preliminary results from the LLNL analysis.

The cumulative yields recommended by LANL for the four nuclides considered are in agreement with the projections from the isotope dilution experiments within essentially ± 1σ. We do not see a significant difference in the projected yields from our analyses with quadratic or linear fits to the data from the isotope dilution experiments. The uncertainties of the ratios of the LANL recommendations to those we project are, as far as we can see, about ± 3%.

Table 5a. Comparison of projected cumulative yields from the analysis of isotopic dilution experiments with those recommended by LANL[19].

Nuclide	LANL	Analysis I	LANL / Anal. I	Analysis II	LANL/Anal. II
^{95}Zr	4.76 ± 0.14	4.75 ± 0.07 [c]	1.002 ± 0.033	-	
^{99}Mo	6.20 ± 0.14	6.22 ± 0.12 [d]	0.997 ± 0.030	-	
^{144}Ce	3.66 ± 0.11	3.66 ± 0.05[a,e]	0.999 ± 0.032	3.67 ± 0.05[b,e]	0.999 ± 0.033
^{147}Nd	2.08 ± 0.05	2.13 ± 0.04[a, f]	0.976 ± 0.028	2.11 ± 0.03[b, f]	0.986 ± 0.028

[a] Assumes the quadratic fits shown in Figures 3 and 15.
[b] Assumes that the data from the isotope dilution experiments are best described by linear variations with average neutron energy.
[c] ^{95}Zr - estimated from the average relative abundance of ^{95}Mo and the average elemental yield of Mo, both taken as independent of energy.
[d] ^{99}Mo - estimated from the average isotopic abundances of ^{98}Mo and ^{100}Mo and the average elemental yield of Mo taken to be independent of energy.
[e] ^{144}Ce - estimated from the energy dependence of the ratio $a(A = 144, E_n) / a(A = 144, th)$ for ^{144}Nd and the average elemental yield of Nd taken to be independent of energy.
[f] ^{147}Nd - estimated from the average elemental yield of Nd and the average of the isotopic abundances of ^{146}Nd and ^{148}Nd.

Table 5b. Comparison of projected cumulative yields from the analysis of isotopic dilution experiments with preliminary evaluations of LLNL[20, 21]. See footnotes to Table 5a.

Nuclide	LLNL	Analysis I	LLNL / Anal. I	Analysis II	LLNL/Anal. II
^{95}Zr	4.77 ± 0.03	4.75 ± 0.07 [c]	1.004 ± 0.016	-	
^{99}Mo	6.12 ± 0.08	6.22 ± 0.12 [d]	0.984 ± 0.024	-	
^{144}Ce	3.69 ± 0.02	3.66 ± 0.05[a,e]	1.008 ± 0.015	3.67 ± 0.05[b,e]	1.005 ± 0.016
^{147}Nd	2.07 ± 0.04	2.13 ± 0.04[a,f]	0.972 ± 0.026	2.11 ± 0.03[b,f]	0.981 ± 0.023

Assuming that the cumulative yields recommended by LLNL can be taken as those estimated for an average neutron energy of 1.5 MeV, all are in good agreement with our projections. In summary, it appears that the three sources of data evaluation on the cumulative yields of ^{95}Zr, ^{99}Mo, ^{144}Ce and ^{147}Nd are in agreement at an average neutron energy of 1.5 MeV.

3. Projection of Yields to $< E_n > = 1.9$ MeV

In our opinion, projection of the cumulative yields of all four nuclides to the conditions of the Jezebel critical assembly neutron distribution cannot be settled satisfactorily without additional, highly-accurate experiments. Our fit to the cumulative yield data shown in Figure 14 from the data provided by Chadwick[18] provides an estimate of 2.11 ± 0.05% for the cumulative yield of ^{147}Nd, the preliminary fit to the LLNL[21] data gives 2.11 ± 0.05%, and a linear fit to the isotope dilution data of Figure 3 gives 2.12 ± 0.03%. However, the quadratic fits shown in Figure 3 would suggest a yield of ~ 2.16%.

The implication that the elemental yield of Zr increases with energy (Figure 10) suggests that at some point, energy dependence for mass chains in the vicinity of A = 95 - 100 might be found. The scatter in both the isotopic abundance and cumulative yield data for ^{95}Zr from the isotope dilution experiments makes any projection very uncertain. We note that Chadwick[15] has also tentatively concluded that there may be significant energy dependence for the ^{95}Zr yield. Notwithstanding the apparent constancy of the yield of ^{99}Mo, if the Zr elemental yield continues to increase with energy, some energy dependence of the ^{99}Mo yield might also become evident. Finally, the data shown in Figures 1 and 4 imply that energy dependence of ^{144}Ce is likely.

The table does not discuss yields for ^{155}Eu or ^{156}Eu because they were not measured in the isotope dilution experiments. We have found no relatively recent reports on cumulative yields of these isotopes in the open literature. The only yields of which we are aware are those reported by LLNL and we take these as reasonable representations of the current state of knowledge[11].

4. Conclusions

In the main, the analyses of data from the recent studies by LLNL, LANL and that presented here are in good agreement with respect to the cumulative fission yields of the

principal nuclides used for weapons diagnostics and nuclear detonations in general over at least to an average neutron energy of 1.5 MeV. The yields appear to be known at this energy to uncertainties of (3-5)%. Radiochemical results are mainly from experiments in critical assemblies while all of the isotope dilution experiments came from reactor irradiations. The uncertainties on yields are likely dominated by correlated systematic uncertainties that are difficult to estimate and possibly unknown. Achieving overall uncertainties of 2.5% will require several experiments with ~1% measurement precision so that the presence of correlated and unknown uncertainties at the 2.5% level can be evaluated.

We conclude that

a. The isotopic and elemental yields produced by the isotope dilution methodology must be considered excellent on the average.

b. The data sets from the three evaluations agree at the level of ± 1σ or a relative error of about ± (3-4)%.

c. There are very likely small energy-dependences in the cumulative yields of ^{144}Ce and ^{147}Nd but this conclusion is not yet completely quantified. This is especially important when one wishes to provide cumulative yields at ~ 1.9 MeV with 1σ uncertainties of 2.5%.

d. The energy dependency of the elemental yield of Zr seems evident.

e. From the point of view of understanding the energy dependence of fission yields in general, a method such as isotope dilution or some other form of mass spectrometry should be considered seriously. The ability to obtain isotopic abundances with very small errors provides measurements that are not available with any other method. They can, in principle, yield a fairly broad range in mass number of high-quality cumulative yields for comparison with theory.

V1. Future Studies

1. Theoretical

1.1. Introduction

While very detailed in-depth reviews have been undertaken at both LLNL and LANL, it seems unlikely that definitive conclusions on the energy dependence of specific fragment yields can be established to the desired accuracy without future experimental and theoretical studies. The most accurate and extensive data outside of the critical assembly studies are those of Maeck and coworkers that have been discussed in detail in the previous sections. However, these results and the energy dependencies that they measure, while appropriate for reactor design, are in the region 200 to 1300 keV average neutron

energy that is significantly below the energy region of interest here ($< E_n > \sim 1.9$ MeV). A simple extrapolation using accurate thermal data, the results from isotope dilution experiments and the higher-energy measurements, with their estimated uncertainties of 4-5%, does not seem likely to produce results with a high degree of confidence unless improvements are made in the theoretical understanding of fragment production and new, more accurate experimental measurements are performed.

On the theoretical side it is well known that in fission of the actinides, the heavy mass peak is anchored by the Z = 50, N = 82 doubly-magic shell and that the stability afforded by this shell is lost rather slowly with increasing excitation energy. In addition to this major shell there are other possible sub-shell structures in the heavy fragment and in the complementary light fragment. Qualitatively, the scission process proceeds with the heavy fragment remaining almost spherical and the light fragment deforming enough so that a charge separation point can be reached where the Coulomb repulsion can overcome the nuclear surface tension and scission occurs. In cases of interest here it is believed that scission occurs at relatively low excitation energy. Following scission, however, the two fragments are excited by the dissipation of their surface tension (deformation) energy as they reconfigure to optimum shapes. The two now separate fragments are excited in energy with the resulting emission of 2-3 neutrons. The excitation energy and subsequent neutron emission comes mostly from the light fragment that possessed the major part of the deformation energy (i.e., neck) at scission. As the excitation energy at scission is increased more phase space can become available for the emission of fragments with Z,A deviating from the doubly-magic 50,82 and the heavy mass peak along with the complementary light mass peak will tend to broaden especially in the lower-yield tails that are of interest here. Thus one would expect these tail yields to increase slowly with energy.

A separate phenomenon is the rise of the symmetric fission component, which is believed to come from a separate dynamical path to scission with the relative probability of the symmetric or asymmetric mode being determined at the fission barrier. Thus the symmetric to asymmetric competition seems to be mainly an effect of the excitation energy available at the barrier while the broadening of the asymmetric (or symmetric) peaks appears to be a phenomenon more associated with scission. An extreme model developed by Wilkins et al.[22] assumes statistical equilibrium at the scission point and then estimates relative mass and charge division at scission based on a multidimensional potential energy surface where shell and pairing effects have been included explicitly. This model was moderately successful in describing the overall pattern of mass yields throughout the whole actinide region. It is, however, not accurate enough to directly tackle the peak broadening effects of interest here. Nevertheless, one can imagine a semi-empirical adaptation of this approach that might be useful for the current problem.

As more excitation energy (and phase space) becomes available the shells and sub-shells become relatively less important, the symmetric yield increases and the high mass tails broaden. The excitation energy required for this change is generally some 10s of MeV and there are many issues that limit a truly quantitative modeling of this process.

In addition to the effects from the high binding energy of closed shells, it is known that the heavy fragment mass peak shows significant fine structure that is believed to be due to the extra binding energy available when an even-even fissioning nucleus divides into two even-even fragments and approximately 2 prompt neutrons. This effect is most prominent in the heavy peak because at scission the shell keeps the heavy fragment as near spherical as possible and the light fragment, being highly deformed, receives most of the excitation energy. Thus, the pairing effects on the light fragment are more likely to be obscured by de-excitation and prompt neutron emission. What happens in the regions of highly asymmetric splits, where the heavy mass fragment is significantly beyond the peak yield is not completely clear. Detailed studies of mass distributions for thermal neutron fission have shown complex fine structure especially in the heavy mass peak and some of that is probably due to the additional binding present in even-even systems. In particular, the ratios of specific mass yields for approximately 600-keV neutron fission to thermal neutron fission from Maeck results show, particularly in the region of the heavy peak, an odd-even effect that is consistent with the "washing out" of the pairing stability. In this case the odd A systems are rising more rapidly that neighboring even masses by 3-5%. This is typical of the pairing effect being overcome and the energy scale for such a process would be expected to be similar to the pairing gaps of about 2 MeV. Results shown by Maeck show fine structure more clearly and further into the heavy mass tail for the n + ^{235}U fission reaction than for n + ^{239}Pu reaction. Whether this is due to higher-quality ^{235}U fission data or because after thermal capture ^{240}Pu is at roughly 0.5 MeV higher excitation energy above the fission barrier than is ^{236}U is not known.

The presence of two major nuclear structure effects, the Z = 50 N = 82 shell and the pairing effects, suggests that a simple linear dependence of a particular fragment yield on energy is unlikely to be correct. The effects from the smaller pairing energies tend to be lost more rapidly with increasing excitation energy as compared to the much larger effects from the stability of the doubly-closed shell. In addition there could be small differences in the fragment mass spectra for specific spin and parity states since these quantum numbers will be preserved through the whole process. Such effects should be lost rapidly as more channels open in the fission process. However, s-wave thermal fission may be a special case especially in n + ^{239}Pu fission where the fission proceeds predominately though only the 0^+ state because there are no 1^+ states readily available at the fission saddle below the pairing gap. This suggests that one should be wary of using thermal data to anchor a smooth function describing the energy dependence for specific fragment yields.

In the sections below we will discuss broadly some possibilities for future theoretical and experimental initiatives that might be useful in obtaining a more quantitative picture of the energy dependence of isotopic yields for specific fission fragments. On the theoretical side an empirical approach using a statistical scission point model might be useful in defining shapes and expected magnitudes for the energy dependence of specific fission fragments. Experimentally, there are two directions that seem to hold some promise. The first would use a high current accelerator to produce proton and deuteron beams to obtain neutrons from (d,d) and (p,^{6}Li) reactions at a variety of neutron energies. High currents could allow direct measurements of fragment yields using radiochemical or

isotope dilution mass spectrometry. These experiments measure the cumulative yields at or near the end of the β-decay chains. A second set of experiments would look at the primary fragments before β decay. A direct reaction correlation experiment could use the ^{239}Pu(d,p) reaction to excite ^{240}Pu at a spectrum of known excitation energies and then detect γ rays in coincidence from specific prompt fission nuclides to measure the energy dependence for the formation of these nuclides. Fragments from the n + ^{239}Pu reaction driven by a variable energy neutron source could also be observed directly by a time projection chamber where the fragment trajectories are bent and analyzed with sufficient precision to separate individual mass components. Both of these experiments involve studying fragments from reactions where fission has already occurred so there is no need for an independent fission flux monitor.

1.2. Theoretical Modeling

The Maeck data on energy dependences for fission of ^{239}Pu and ^{235}U appear to show pronounced odd-even effects that are presumably due to the increased binding for paired nuclear configurations. In general the ratios of yields of specific fragments show definite odd-even fluctuations when data at neutron energies of a few hundred keV are compared to thermal neutron data. The odd A fragments show a larger change in yield than neighboring even A fragments. The Maeck data for ^{239}Pu show effects between neighboring masses of order 5% whereas for the data from ^{235}U fission, a more regular structure is seen but only with about half the magnitude. This is the result to be expected if original pairing fine structure were washing out and the yields of odd A nuclides were catching up to those of neighboring even A. It is not at all clear how this healing will proceed as the energy is increased to the 1-3 MeV range. Therefore, even though both the thermal and Maeck data may be accurate to 1-2% it would seem that any extrapolation to higher energy would be problematic until the washing out of the pairing fine structure is understood.

A relatively straightforward statistical model might be used to estimate the effect of the increased pairing stability as the total excitation energy available to the fragment is increased using level densities that include both shell and pairing effects. In these level densities the suppression of the ground state energy for a paired system results in a relative decrease in the low-lying levels for excitations below the pairing gap. A neighboring odd nucleus will not be quite as strongly bound but it will have more low-lying excited states available. If the excitation energy is increased for these two systems then the odd system starts from an effectively high potential energy but as energy is added the increasing rate of available levels allows it to catch up to the even system at some energy that is around the pairing gap (order 2 MeV). At this excitation energy the even-even configuration is no longer preferred over the odd-odd configuration and the pairing effects are effectively washed out. A similar approach applies to the shell corrections except that the relevant energy scale is now on the order of some 10s of MeV. To apply such a concept to the emission of fragments at scission requires an assumption that statistical effects outweigh any dynamic effects that may be present. This approach was shown to produce gross properties of fission by Wilkins et al.[22] and is consistent with

our picture of fission that indicates that the descent from saddle to scission is a relatively slow viscous process.

In the present case if one took an empirical potential energy surface at scission that described a smoothed mass distribution for the emitted fragments then it might be possible to introduce the pairing energy fluctuations and then quantitatively describe the fragment mass distribution and its evolution with increasing excitation energy. A similar approach was taken in the scission point model described by Wilkins et al.[22]

Effectively smoothed prompt fission distributions are known from correlation experiments that measure the energies of the two coincident fragments. Data are available for thermal and higher-energy neutrons interacting with ^{235}U and ^{239}Pu. Then using microscopic level densities the excitation energy of the system could be raised and the relative mass of each fragment would come from its total phase space (i.e., summed level density). How quantitative this process might be remains to be seen but this is not a very big or demanding project. A simpler project would be to look at the ratios of the sum of the total available levels for some of the fragments of current interest to a few fragments at the peak of the heavy mass peak where it is known that there is little or no energy dependence in the yields. Either of these approaches should give, at a minimum, information on the expected shape of the excitation function for those nuclides.

A complication of this approach is that what is obtained is a prompt mass (and charge) distribution after prompt neutron emission but before β decay. The calculated prompt distribution would need to be evolved to give the cumulative distribution that is of interest here. This procedure seems to be relatively straightforward.

2. Experimental

In order to establish the parameters for the energy dependence of specific fragment yields to the accuracy and precision required, it might be necessary to perform new experiments. Ideally it would be desired that all required data be collected from a single experiment where the energy is varied up to 3 - 6 MeV incident neutron energy and individual fragment yields measured to 1% accuracy. Because the major source of systematic uncertainty is in the determination of the number of fissions, any new experiment must address this issue. In fact, serious attempts to reduce errors should probably encompass several approaches simultaneously to help shed light on the sources and magnitudes of systematic uncertainties.

It is convenient to distinguish two broad classes of experiments, those that measure chain yields post irradiation and those that measure chain yields directly (after prompt neutron emission but before β-decay occurs). For each class, several analysis techniques can be employed to determine the yield of a specific fragment. Four experimental approaches are considered below in more detail: (1) isotope dilution mass spectrometry, (2) post-irradiation γ- and β-decay spectroscopy, (3) prompt mass spectrometry, and (4) prompt γ-fission coincidence spectroscopy with direct reactions. It seems clear that in most cases

better experimental insight will require an intense mono-energetic neutron source. The basic considerations for such a facility are also discussed.

Post-irradiation decay spectroscopy

Any new experimental program must include post-irradiation decay spectroscopy. Experimentalists should aim to uncover and understand systematic uncertainties inherent in the β- and γ-ray spectroscopy methods used by LLNL and LANL during underground testing. As with isotope dilution, very precise measurements of the energy dependence of the yield of *specific* isotopes can be made with this technique, probably to 1%. There are systematic limitations in our ability to fit γ-ray peaks and β-decay curves that probably limit this technique to at best 0.5% systematic uncertainty. A precision measurement today would require an appropriate neutron facility (~5×10^6 n cm^{-2} s^{-1}) coupled to the requisite radiochemistry and accelerator mass spectroscopy facilities. By comparing yields at different irradiation energies, uncertainties in detector efficiencies and branching ratios are substantially reduced, but many detailed issues in determining the target masses, number of fissions, spatial, temporal, and energy variations in the neutron flux, etc., will need to be addressed. In particular, these measurements require a monitor for the number of fissions that is not sensitive to the incident neutron energy. A fission chamber might be appropriate, but energy-dependent effects will have to be studied. Reducing the *absolute* fragment yield uncertainties with this technique to ~1% is considerably more difficult. The inherent difficulty is the accurate determination of the absolute number of fissions. As discussed earlier, fission chambers, which have been used extensively, have normalization uncertainties of ~1.4%. Time Projection Chambers (see below) or some other more informative monitoring system that can correct for energy-dependent efficiency effects would be able to reduce this uncertainty. Another approach might be to measure all mass chains as a way to determine the number of fissions, but this would be a very large effort and not warranted unless necessary.

Isotope dilution mass spectrometry

The technique of isotope dilution has the lowest uncertainties of all techniques that have been employed to date. Isotopic ratios are measured today with an accuracy of 0.1% for sample sizes of 10^{10} atoms or less. The yields for every stable and long-lived isotope of an element are determined within the same experiment. These measurements would have uncertainties of 1% or less. Many of the issues associated with this technique are shared with decay spectroscopy, but isotope dilution has lower inherent systematic uncertainties. For *absolute* measurements, isotope dilution also benefits from not needing precise decay branching ratio information.

Prompt mass spectrometry

A very appealing approach to a precise measurement of mass chain yields is to directly measure the mass of both fission fragments in coincidence with a fission event via electromagnetic separation. Several groups have performed prompt isotope yield measurements via mass spectrometry; the most modern results come from the Lohengrin spectrometer at the Institute Laue-Longevin in Grenoble[23]. While these experiments have been of enormous scientific value, they have not been of sufficient accuracy to determine chain yields due mainly to systematic uncertainties in determining the

efficiency of the spectrometer which is ~5% and which is dependent on the kinetic energy of the fragments and other variables. A program to determine the efficiency of the spectrometer seems plausible**.**

Ideally, a mass spectrometer with a large angular and energy acceptance would improve the situation substantially. In such a case, there is no need for a separate fission monitor, because the fragments are recorded for each fission event. The yield is simply the number of fragments with mass A divided by the number of events. Corrections for possible biases toward specific fission events are small if the overall efficiency for detecting a fission is ≥ 90%. The fission Time Projection Chamber (TPC) being developed by the Neutron Induced Fission Fragment Tracking Experiment collaboration is promising in this regard as each fission event is recorded with high efficiency (~98%). The track for each fragment is recorded as it slows down in a gas chamber with spatial resolutions of a few mm and the energy loss along the path length is determined. While the current fission TPC has been designed for precision (1%) cross-section measurements and has a mass resolution of a few nucleons, a design with the necessary mass-to-charge resolution might be possible using an external magnetic field. If such a design can be achieved, a TPC not only can serve as an improved fission chamber for determining the number of fissions, but also can directly measure the prompt fragment yields for specific mass chains with high accuracy. The capital costs for such a detector system are unclear without further study, but if the fission TPC design can be leveraged, the capital costs are plausibly ~$2M. However, it should be noted that a precision measurement with this technique requires a rather intense mono-energetic neutron source capability (~5×10^8 n/cm^2/s) not currently available in the US.

Direct reaction γ-fission correlation experiments

Fission probabilities, fission barriers, and surrogate (n,f) cross sections have been measured for a large number of actinides using direct reaction correlation techniques. This technique involves observing fission from a residual system that has been excited by a direct reaction to an energy above the fission barrier. An example would be fission of ^{240}Pu following an excitation by the ^{239}Pu (d,p) reaction. Observing the energy of the outgoing proton leads directly to the excitation energy for ^{240}Pu for that event. By measuring as a function of proton energy it is possible to get an excitation function for the probability of fission.

A possible expansion of this concept would be to also measure γ rays characteristic of specific fission fragments in coincidence with the proton and fission fragment. Then the relative yield of specific fragments versus excitation energy could be obtained from the quantity $R[IY(Z,A)] = N_\gamma(Z,A) / N_f$, where R[IY(Z,A)] is the relative yield of a fission fragment, $N_\gamma(Z,A)$ is the measured intensity of a γ ray emitted by a fragment and N_f is the number of fissions recorded. All quantities are measured directly and the whole excitation function for $0 \le E_n \le$ 5-6 Mev is measured at once. Then it would be a question of determining the absolute intensity $I_\gamma(Z,A)$ for the particular characteristic γ ray assayed.

In order to get a reasonable count rate for this experiment it is important to achieve as high efficiencies as possible for the fission and γ-ray detectors. This could be done with a setup similar to what was used previously at LANL.[24] This whole system can be quite compact and should be able to fit inside a 4π γ-detector array such as GAMMASPHERE or a more modest array. To obtain the absolute value for R[IY(Z,A)], all that is needed is to determine the total fragment yield for a particular nuclide from one or a few measured γ rays using known level systematics for that nuclide. It should be possible to determine yields fairly accurately and most factors do not change rapidly with energy. Thus, the relative energy dependence should be dependent primarily on counting statistics and could be quite accurate. Note that the absolute yield per fission as a function of energy is not dependent on the solid angles of the proton or fission detectors. Because events are measured in coincidence, only those corresponding to a measured proton and fission fragment are collected.

This experiment could be performed with existing tandem accelerators or with the LBL 88-Inch Cyclotron. No neutron source is required. A relatively simple test experiment could be performed to evaluate empirically whether this technique could provide a measurement to the requested accuracy.

A good experimental group with experience at GAMMASPHERE, for example, should be able to perform an absolute measurement of R[IY(Z,A)] to an uncertainty of 5% or less and a relative energy dependence to even smaller uncertainty. If this type of experiment were coupled with a microscopic theoretical investigation of the fission process it would prove very interesting.

Probably the largest uncertainty will be associated with the determination of absolute individual fragment yields from the observation of a few known characteristic γ rays. Uncertainties would likely be less serious in the measurement of the relative excitation function for a specific nuclide. Using standard techniques it should be possible to measure proton energies (and thus the residual excitation energies in the product nucleus) with a resolution of 50-100 keV, which seems adequate for the current problem.

Mono-energetic neutron source requirements

We are not aware of a mono-energetic neutron source facility in the United States where it is feasible to perform the experiments outlined above. The traditional radiochemistry methods require ~5×10^{6} n cm^{-2} s^{-1} at several energies and the large acceptance mass spectrometry measurements require ~5×10^{8} n cm^{-2} s^{-1}. Similarly, adequate critical mass facilities are no longer operating in the country and there is still the problem that results are obtained as averages over a broad energy range. Reactor-based isotope dilution and radiochemistry results have been performed to the required accuracy but the energy range available is inadequate to completely tackle the problem of understanding the energy dependence of fragment yields over the energy range of interest. It is not clear how this technique could be expanded to the 1-3 MeV range in neutron energies.

The technologies for mono-energetic neutron sources of the required intensities are well established, and LLNL and LANL both have the facility space to house such a capability.

The capital costs to develop this capability are probably ~$3M in order to acquire or refurbish an ion source, accelerator, beam transport, production targets, diagnostics, and control systems. At LLNL, for example, Larry Ahle has proposed such a capability (named ALEXIS) to be housed in B194 that can provide neutrons within 1-2 years. Current plans project additional costs of ~$2M to provide ~$10^7$ n s^{-1} in the 0.1-0.4 MeV region using the ^{7}Li(p,n) reaction and ~10^9 n s^{-1} in the 5.0-9.0 MeV region using the d(d,n) reaction. An intense neutron source in the energy range 1.0-5.0 MeV is most easily provided by the t(p,n) reaction, which requires a tritium target. A project plan would need to be developed for a tritium target capability at ALEXIS, but the targets are available commercially and the safety basis for B194 allows such targets. The ALEXIS proposal utilizes an existing 3-MeV Pelletron accelerator, but Radio Frequency Quadrupole (RFQ) accelerators are possible at a cost of ~$1M and could be used as the basis for such a facility as well.

Additional Comments on Data from Radiochemical Studies

Because modern measurements on underground test debris were made using high-resolution γ-ray detectors, new benchmark radiochemical measurements of cumulative yields could be applied to re-evaluate the debris data and substantially reduce systematic errors associated with the older radiochemical measurements. The accuracy of underground test debris analyses has been a concern since the beginning of underground testing. We refer the reader to two recent references[25,26] that address the wide variety of errors connected with the analyses and interpretation of the radiochemical analyses of underground test debris. While radiochemical measurements probably cannot attain the same precision as mass spectrometric measurements, we note that they can attain experimental precisions of 0.5 to 1% based solely on uncertainties in the determination of full-energy peaks in γ-ray spectra. It should be possible to reduce uncertainties in absolute photon intensities and other nuclear parameters for the few nuclides of interest through new studies of their radioactive decay. Benchmarking against highly-precise mass spectrometric measurements is a second possible route toward reducing uncertainties from these sources as well as uncertainties in detector efficiencies.

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